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Detection of low energy positive ions by scintillation methods

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DETECTION OF LOW ENERGY POSITIVE IONS BY SCINTILLATION METHODS

BY

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Contents

List of Illustrations

List of Tables

Introduction

This thesis deals with the feasibility of efficiently detecting low energy psoitive ions, those with energies of less than several hundred electron volts, using phosphors.

The scintillation technique, first used visually and greatly improved in the last ten years by the use of a photomultiplier tube, has greatly advanced detection in high energy fields. The photomultiplier is capable of delivering milliampere pulses of current of short duration and these can be registered directly by the many electronic devices currently in use.

Extensive work has been done with phosphors which scintillate under high energy bombardment, due in part to their wide use in the television industry, but very little is known of their use for the detection of low energy positive ions.

The main advantage of the scintillation technique for low energy particles lies with the relatively short time constants of the phosphorescent materials. If low energy ions produce scintillation, this process of detection would allow not only the detection of ions but, in conjunction with the techniques of mass spectroscopy, would enable one to distinguish between the types of ions being produced. Furthermore, if low energy positive ions can be detected, there is a possibility that metastable states could also be detected. A method of detecting ions or

metastables of low energy would be a valuable aid in studying the ionization of air and other gases, particularly the photoionization processes of the upper atmosphere.

Interest in the region of the electromagnetic spectrum below 2000 angstroms (the vacuum ultra-violet) is primarily due to the problems concerning the composition of the upper atmosphere and the photo-electric and photo-conductive effects in solids.

Of particular interest in the study of upper atmosphere physics are the mechanisms responsible for the creation of the ionized layers, and a strong interest has developed in the responsible portions of the solar spectrum, the vacuum ultra-violet.

The atmosphere is not a continium, but consists of a number of distinct layers as shown schematically in Appendix A.

Perhaps the most important effect of the solar ultra-violet radiation on the upper atmosphere is the ionization of its constituent gases. The ionization commences from a height of about sixty kilometers and extends up to the highest limits of the atmosphere. These ionized regions which are discussed in Appendix A are known collectively as the ionosphere. To the radio-physicist, the presence of these ion layers in the upper atmosphere, and their mechanism of formation are of prime importance and hence, have been the object of close study for many years. To

the physicist interested in such phenomena as the bombardment of gases by charged particles, electric discharges in rarified gases, ionization of gases and photo-chemical reactions, this region furnishes a natural laboratory in which nature is performing experiments on a scale beyond the scope of any man-made laboratory.

Much of the impetus for research on the upper atmosphere is due to the importance of these ionized layers to radio communication. A thorough knowledge of these layers would enable one to predict the maximum usable radio frequencies.

The absorption of photons having wavelengths in the vacuum ultra-violet and the soft x-ray region is the main mechanism for the formation of ions in the upper atmosphere. At high altitudes the atmosphere consists mainly of O, N, O_2, N_2. The energies required for the ionization and dissociation are relatively high, as indicated in Table 1.[1] The solar radiations reaching the upper

(1) Gaydon, A. G., Dissociation Energies and Spectra of Diatomic Molecules, New York, John Wiley and Sons, 1947, p. 93, p. 97, p. 161.

N_2	$N + N$	9.76 ev	O_2	O_2^+	12.30 ev
N_2	N_2^+	15.57 ev	O	O^+	13.60 ev
O_2	$O + O$	5.08 ev	N	N^+	14.54 ev

Table No. 1. Ionization and Dissociation Energies

atmosphere contain photons of sufficient energy to cause

ionization of these gases, hence the formation of ions in this region.

The formation process for ionized strata may be understood as follows. The atmosphere of the earth is considered to be a single gaseous element, the density of which decreases exponentially upwards.(2) Radiation entering the

(2) Mitra, S. K., The Upper Atmosphere, The Asiatic Society, Calcutta, India, 1952, p. 279.

atmosphere from outside is absorbed in producing ionization, therefore the rate of ion production will be a maximum at a certain level. This is because the rate of absorption at any height is controlled by two factors, the intensity of the incident radiation and the density of the absorbing gas. As the radiation enters the atmosphere from above, its intensity decreases and the density of the absorbing gas increases. These layers of maximum ionization are modified, however, by the rate of recombination of the gases concerned. A theory of the formation of these strata, known as the Chapman Theory, has been formulated and is developed in Appendix B.

The Chapman Theory can lead to some important results. However, among the factors in the equations that are not readily determined are the absorption coefficient per unit mass of the gas and the number of ions produced by absorption of unit quantity of radiation.

More complete data regarding the absorption cross

sections of N_2, O_2, N and O in the extreme ultra-violet are necessary, because the ionizations in the upper atmosphere are controlled by the absorption of these gases.

A vacuum spectrograph which is suited to making such measurements is at the present time being constructed by the Physics Department, Missouri School of Mines and Metallurgy.(3)

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(3) Henderson, J. K., Unpublished Thesis Material

Cross sections can be determined if the number of photons incident per second on a given mass of gas and the number of ions per second produced are known. The strength of the ion current is then a direct measure of the ionization efficiency. One of the main problems associated with the use of ionization chambers is the measurement of small ion currents. These ion currents run from 10^{-4} to 10^{-15} ampere. Once measured these currents give the sum results of the processes that have taken place but no insight into the processes individually. For example, in measuring cross sections for oxygen, it would be valuable to know if the current was primarily O_2^+ ions or O^+ ions, how many atoms were left in metastable states and similar information.

A technique which might be valuable in such measurements is the time resolution mass spectrograph method. A sketch of a possible set-up is shown in Fig. 1. A suitable phosphor, coated on conducting glass forms the top. Photomultiplier tubes are placed in direct contact with

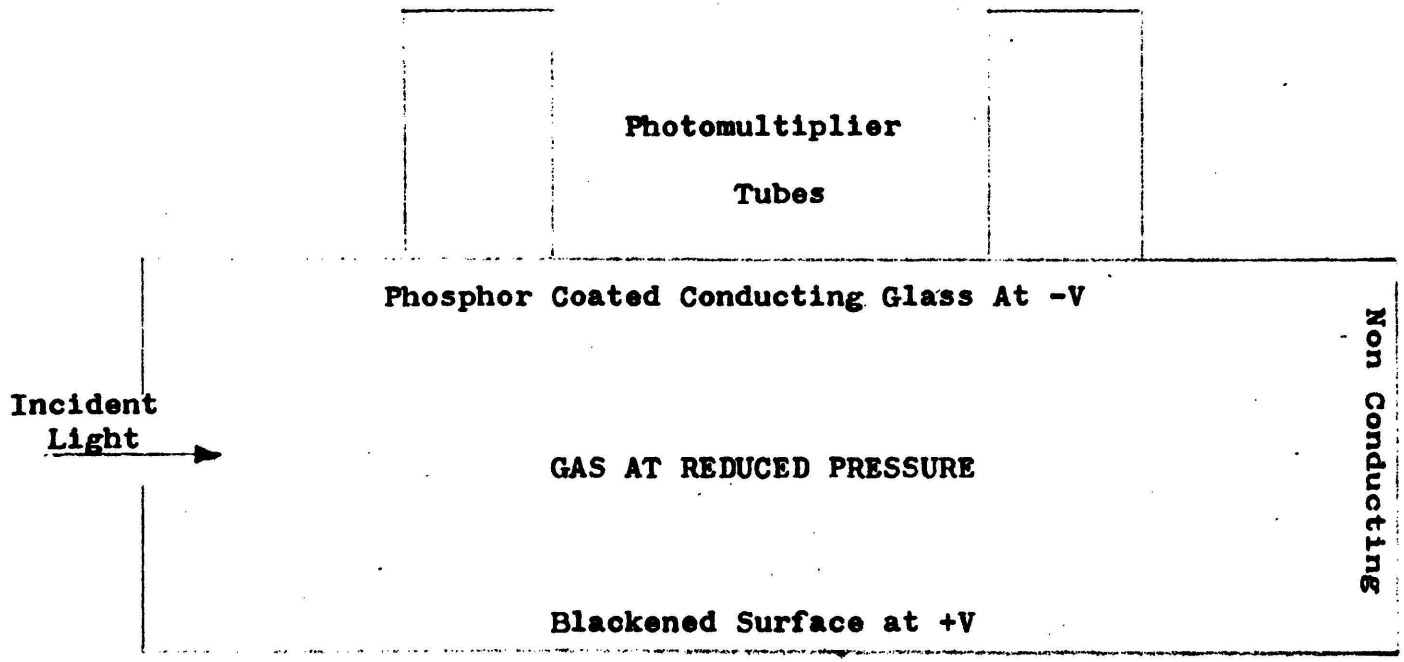

Fig. 1. A possible set-up for using phosphor in a time resolution mass spectrograph method.

the glass surface and a low voltage is placed between the upper and lower plates. The incident photons, which ionize the gas in the chamber, are pulsed at a rate such as to allow the ions produced by earlier pulses to be swept out by the electric field before arrival. The time of incidence of the ions on the phosphor will depend upon their masses and the interval between the arrival of ions of different types would be short. Any device used to measure these ion currents must have an equally short time response and must detect small currents. Phosphors coupled with photomultiplier tubes meet these requirements.

Instruments which are in general use for measuring small currents such as microammeters and galvanometers usually have a time response much too long for such measurements. Specialized instruments such as the vibrating capacitor electrometer have been developed to measure small currents with a time response on the order of milliseconds but require specialized skill for adjustment.

The field of low energy excitation of phosphors by positive ions is relatively unexplored, and it is possible that additional information on the mechanisms involved in the process may be obtained. These in turn may provide data on the band structure of insulators regarding which there is very little information available at the present time. It is found also that the operating characteristics of a particular phosphor are very dependent upon the methods of preparation and the conditions of use, and this field

can be further explored in this research.

It is hoped that the scintillation technique will provide a simple, accurate method of measuring small ion currents with a short time constant and possibly contribute to the understanding of the mechanisms of the luminescent process.

Luminescence

Luminescence is a general term which includes the many phenomena involving the absorption of energy by a substance and its re-emission as visible or near-visible light.

The first scientific treatment of luminexcence came in the middle of the nineteenth century by Edward Becquerel.[4] He measured the wavelength of the incident radiation exciting the material and the emitted radiation, the duration of the afterglow, and the influence of temperature. Becquerel investigated the luminescence of many materials but it was left to Lenard[5] at the close of the nineteenth century to find that nearly all the mineral phosphors owe their luminescence to the presence of impurities.

(4) Becquerel, E., Ann. Chim. Phys., 22, 244, (1848)

(5) Lenard, P., und Klatt, V., Ann. Phys., 105, 286, (1904)

Since light is a form of energy, energy must be supplied to every system serving as a source of light. The most common method of supplying energy is to heat the system. This is an example of increasing the energy of all the molecules of a system until thermal radiation is obtained, as in a light bulb.

It is possible, however, to transfer energy only to

those parts of the molecule which are responsible for light emission. The molecules can be brought into excited states without increasing their average kinetic energy or without heating the system. For light emitted under such conditions Wiedemann[6] has introduced the

(6) Wiedemann, E., Wiedemann's Ann., 37, 177, (1889)

term luminescence. Luminescence can be divided into two main types, with regard to the mode of excitation. Photoluminescence produced by the absorption of light and electroluminescence, which is produced by the impact of electrically charged particles such as electrons or ions. It is mainly with electroluminescence this thesis is concerned.

Luminescence can be either fluorescence or phosphorescence, depending on whether light emission continues a measurable time after the end of the excitation process. Fluorescence is due to the spontaneous allowed transition of a molecule from an excited state to a lower energy level. The mean life of the process depends only on the transition probability, it is in most cases very short, less than or equal to 10^{-7} seconds and furthermore it is practically independent of temperature.[7] The character-

(7) Fonda, G. K., J. Appl. Phys., 10, 408, (1939)

istic feature of phosphorescence is that a fraction of the excited molecules do not immediately begin to emit light by returning from the excited state to the ground state, but pass instead into a metastable state of somewhat smaller energy than the excited state. From the metastable state the molecules are forbidden the transition to the ground state and can only return to the excited state, with subsequent light emission accompanying the passage from excited to ground state, when the missing energy is restored to them by the heat movement of the surrounding medium. These two processes are shown schematically in Fig. 2.[8]

(8) Pringshem, Peter, and Vogel, M., Luminescence of Liquids and Solids, Interscience Publishers, New York, 1943, p. 11

In an insulating solid, such as those with which this thesis is concerned, the electrons of the lattice atoms are not localized on the atoms but are shared by the whole crystal.[9] The energies of these electrons fall

(9) Garlick, G. F. J., Adv. in Electronics, 2, 168, (1950)

into bands of permitted energy states separated by bands of forbidden states. In an insulator the bands are either completely filled or completely empty. Electrons can be raised above the forbidden band to the conduction band by suitable energy absorption. The removal of electrons from the filled band creates positive holes there that play

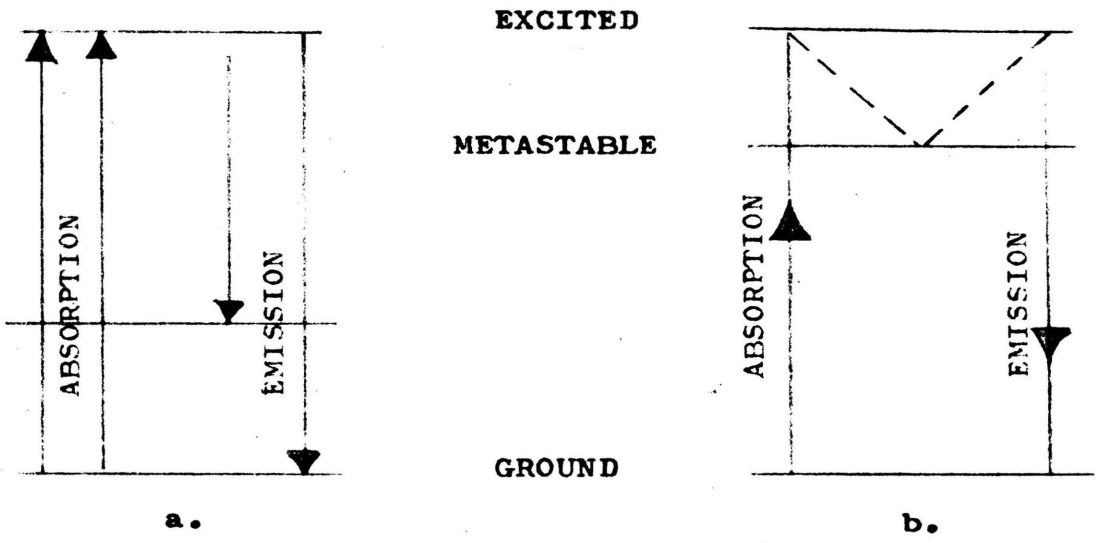

Fig. 2. Energy level diagrams showing the mechanisms involved in a.) Fluorescence b.) Phosphorescence.

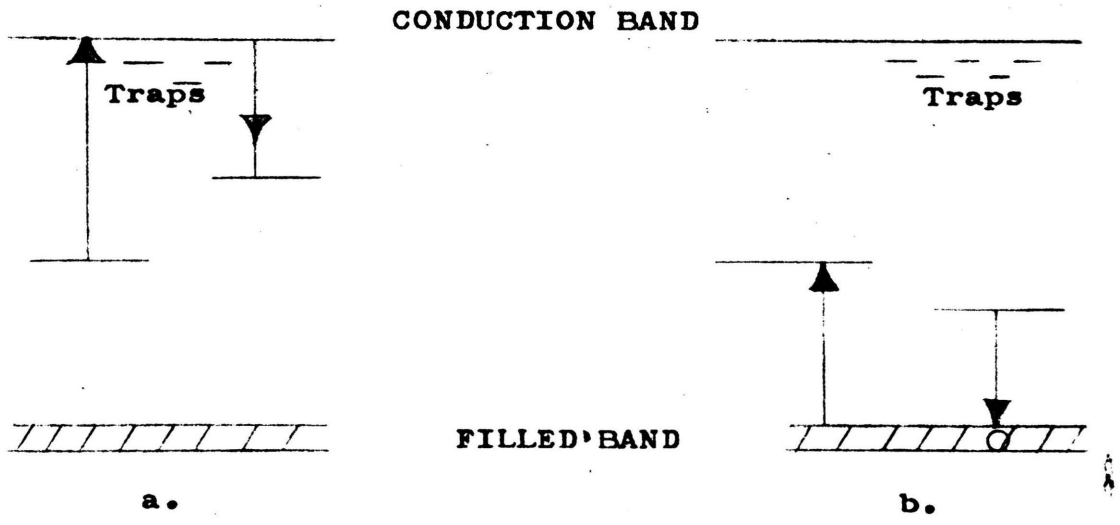

Fig. 3. Diagrams showing how impurity levels may act as donors or acceptors. In a.) impurity acts as donor to conduction band, while in b.) impurity supplies an electron for a hole in the filled band. Electron traps are shown lying just below the conduction band.

a part in the luminescence process.

In phosphors and in most crystalline materials the lattice is far from perfect and its defects give rise to localized energy states for electrons. These may lie in the forbidden energy region. When such levels occur near the conduction band they can capture electrons; therefore they are known as electron traps and are shown on the diagram in Fig. 3. Electron traps give rise to the phosphorescent and thermoluminescent characteristics of phosphors since they delay the emission process by their storage of electrons and the subsequent release of these electrons by thermal agitation. Optical stimulation of the phosphor with visible light may also raise the electrons from traps into the conduction band. Although the possibility exists for transitions from the traps directly to the luminescence centers or the filled band no experimental evidence has been found for transitions directly from the trapping levels.(10)

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(10) Bube, R. H., Phys. Rev., 90, 79, (1953)

Phosphors consist of a host crystal and a consciously added impurity called the activator. Activators may be substitutional atoms, replacing a regular atom of the host crystal, inside the host crystal in interstitial sites, or on surface sites. Phosphors are designated by stating the host crystal followed by the impurity such as ZnS:Cu.

These activators in many phosphors provide localized levels for electrons which may lie in the forbidden region. Because of their position they are of great importance in the luminescent process. These centers may act as donor or acceptor levels in the process as shown in Fig. 3.(11)

(11) Garlick, G. F. J., Luminescent Materials, Clarendon Press, Oxford, 1949

Certain impurities such as nickel in phosphors can also act as "killer" levels. These are fatal for luminescence for they may absorb primary excitation energy directly and convert it into heat or interfere with the luminescence centers and traps such as to increase the probability of non-radiative transitions.(12)

(12) Leverenz, H. A., Luminescence of Solids, John Wiley and Sons, New York, 1950, p. 337

The excitation process in electroluminescence is much more complex than that for photoluminescence. The incident charged particles interact with all the atoms or ions along their path. Particles penetrating into a phosphor will have a range of penetration dependent upon their initial energy. Whereas for ultraviolet radiation, one quantum produces one excited electron, the primary particles incident upon the phosphor will produce many secondary electrons along their paths in the solid.

The important feature of the energy transfer process

seems to be that enough energy be transferred to the electrons in the filled band to produce a hole by exciting electrons to the conduction band. In some phosphors of interest there is a high probability of a considerable number of electrons in the conduction band due to donor levels just below the conduction band. For instance, Hahn[13] has investigated the electrical properties of

(13) Hahn, E. E., J. Appl. Phys., 22, 855, (1951)

zinc oxide and found a high density (10^{17}-10^{18}/cm^3) of donor levels 0.04 ev below the conduction band. This would indicate that luminescence is not wholly dependent upon having electrons in the conduction band.

A model proposed by Lamb and Klick[14] for CdS:Ag

(14) Lamb, J., and Klick, C. C., Phys. Rev., 98, 909, (1955)

provides a satisfactory picture. In this model absorbed energy produces a free hole and an electron in the conduction band. The hole may then capture an electron from a filled luminescent center with the energy dissipated in thermal vibration and an electron in the conduction band can make the transition to the luminescence center producing emission. Thus the center is reset for the next cycle of operation. This model is shown schematically in Fig. 4.

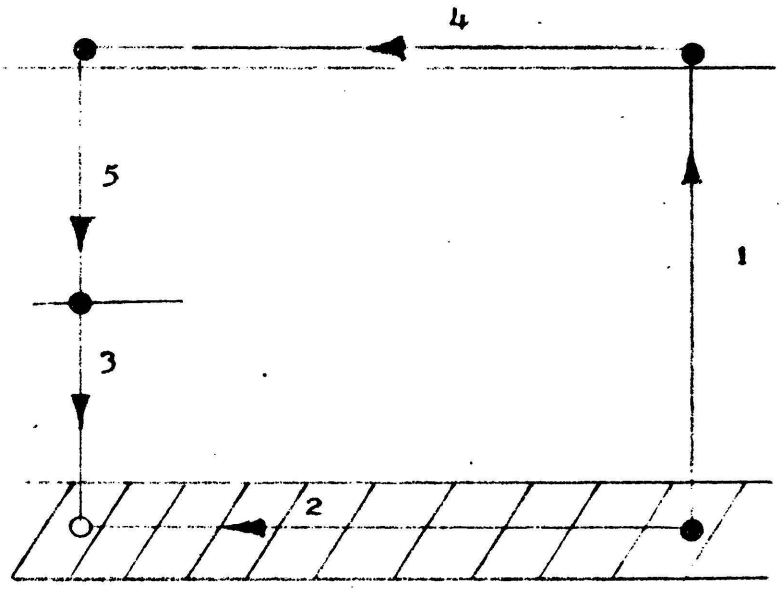

Fig. 4. Lamb and Klick Model

1. Excitation.
2. Hole migration.
3. Capture of Electron from luminescent center by hole.
4. Electron migration.
5. Electron capture resulting in luminescence.

For slow particles there are relatively large surface forces to overcome before penetration is possible. These surface forces give rise to a considerable dead voltage in some cases. Strange and Henderson[15] maintain these

(15) Strange, J. W., and Henderson, S. T., Proc. Roy. Soc., 58, 368, (1946)

dead voltages are due to surface impurities and have used carefully prepared screens which show little or no dead voltage in the cathodoluminescence process.

A general empirical form for emission due to cathodoluminescence is given by:

$$L = k\, f(i)\, (V - V_o)^q \quad (16)$$

(16) Garlick, G. F. J., Luminescent Materials, Clarendon Press, Oxford, 1949, p. 176

where L is the luminescent emission, V is the particle voltage, V_o is the dead voltage, k is a constant for the material, f(i) represents a function of the current and q is another constant for the material with a value lying between one and three. Although this general relationship is verified by most experimental results considerable differences are often obtained for values of q and V_o.

In cases where the phosphor is coated on a non-conducting surface secondary emission from the phosphor crystals is important, especially when the particles bombarding the phosphor are electrons. If the ratio of emitted electrons

to incident electrons falls below a value of one, the phosphor will "charge up" negatively and decrease the energy of the beam.

Phosphor screens often deteriorate due to several factors. One would certainly expect a physical and chemical destruction of the phosphor due to ionic bombardment if the screen were being used to detect ions. Investigations by Seitz,[17] Lark-Horovitz[18] and others on the

(17) Seitz, Frederick, Discussiond of the Faraday Society, 5, 271, (1949)

(18) Lark-Horovitz, K., Semi-Conducting Materials, Butterworth Scientific Publications, London, 1951

disordering of solids by the action of massive particles have shown that ions are capable of introducing new energy levels and changing the electrical characteristics of semiconductors. Experiments indicate that bombardment produces defects which act as donors and acceptors with activation energies larger and distributed over a wider range of energies than those observed with chemical impurities. These defects consist mainly of vacancies and interstitials produced by the passage of the bombarding ions through the material. On the basis of present theory it is probable that such defects are random rather than regular at low energies.

It has been noted[19] that low energy electrons

(19) Garlick, op. cit., p. 188

bombarding phosphors result in a fatigue of the material which produces a decrease in luminescent efficiency. This may be due to an introduction of new energy levels which produce non-radiative transitions since the effect can sometimes be removed by heating the screen. This is suggestive of the annealing process for energy levels produced in radiation damage.[20]

(20) Brown, W. L., Fletcher, R. C., and Wright, K. A., Phys. Rev., 80, 591, (1950)

With regard to the process of electron penetration into the phosphor and the mechanism by which the electrons reach the luminescence centers, experimental studies have supplied very little information. This is more true of the same processes involving positive ions. One reason for this is that many phosphors exist only as microcrystalline powders and therefore have very complex surface conditions for the incident particles.

Leverenz[21] envisions a strictly kinetic process for

(21) Leverenz, H. A., Luminescence of Solids, John Wiley and Sons, New York, 1950, p. 387

the excitation of phosphors by ions. On this basis the luminescent efficiency should vary with the energy of the primary ion in much the same manner as that of cathodo-

luminescence, except that the minimum energy for appreciable luminescence should be considerable larger than that for cathodoluminescence from the same phosphor since the ions penetrate less than electrons with the same primary energy. This is based on the complex surface conditions which limit the processes to the more regular interior of the phosphors.

A fast ion passing through matter can have two quite different effects on the atoms it strikes. In the first place it can make an elastic collision, giving kinetic energy to the recoil atom. The energy which can be transferred is given by:[22]

$$E = \frac{4\, m_1\, m_2\, E_0 \sin^2 A/2}{(m_1 + m_2)^2}$$

(22) Seitz, F., and Koehler, J. S., Solid State Physics, 2 Academic Press, New York, 1956, p. 313

where m_1 is the mass of the incident particle, m_2 is the mass of the struck nucleus, and E_0 is the energy of the incident particle, and A is the angle of deflection in the center of mass coordinate system. We see the energy given the struck atom is a larger fraction of the energy of the incident particle, the more nearly they have the same mass. The relation also shows that in cathodoluminescence the incident electrons will most probably transfer most of their energy to electrons of the lattice exciting luminescence rather than to the ions of the lattice.

Competing with this process is another resulting from the charge on the ion, the incident particle can excite or ionize the electrons of the struck atom, resulting in an inelastic collision in which most of the energy lost by the incident particle is transferred to the removed electron. The rate of energy loss in this type of collision can be shown (23) to depend, among other things, on the

(23) Bethe, H. A., Handbuch der Physik, 24/1, J. Springer, Berlin, 1933, p. 27

square of the charge of the incident particle in analogy to Rutherford scattering.

It is found that fast charged particles passing through matter will increase or decrease their charge, depending on the particle velocity. According to Slater (24)

(24) Slater, J. C., J. Appl. Phys., 22, 248, (1951)

a simple and fairly accurate rule for which electrons will be removed from the fast particle is that all those will be removed whose velocity in their orbits is less than the velocity of the atom and as the atom slows down it picks up electrons reaching a state of equilibrium. Using a simple Bohr picture of the atom the velocity of the last electron in Xenon would be approximately 10^9 cm/sec which would mean that the atom must have an energy of about 400 Mev to loose electrons. According to Seitz (25) the rate

(25) Seitz and Koehler, op. cit., p. 338

of excitation and ionization is proportional to the ratio of the velocity of the ion to that of the bound electron with which it interacts. Thus as the incident particle loses energy, its rate of ionization will decrease.

In collisions between particles of high energy, the time of collision is so short that the nuclei approach and recede from each other in a time short compared with the period of the electrons in their orbits so they have no time to rearrange themselves.[26] But for slower parti-

(26) Slater, op. cit., p. 247

cles the collision takes a longer time allowing the inner electrons to modify their motions and present electronic shielding. This shielding complicates the theory in this range of energy.

According to Knipp and Teller[27] the energy loss of

(27) Knipp, J., and Teller, E., Phys. Rev., 59, 661, (1941)

ions in the region of energy of 10 kev or less is due to collisions with the atom as a whole. This subject has been treated in a general manner by Bohr[28] and Seitz[29] who

(28) Bohr, N., Kgl. Danske. Videnshob. Biol. Medd., 18, 8, (1948)

(29) Seitz, and Koehler, op. cit., p. 315

state that exact quantum mechanical methods must be used to obtain an accurate picture of the energy loss of slow ions.

The extraction of electrons by positive ion bombardment has been studied by a number of people. The mechanisms involved when the energy of the bombarding ion is of the order of a few hundred electron volts is particularly interesting when applied to the problem at hand. Oliphant and Moon[30] have explained the neutralization of positive

(30) Oliphant, J., and Moon, T., Proc. Roy. Soc., A127, 338, (1930)

ions during a collision with a metal surface by the auto-electronic emission into a state of equal energy in the atom of electrons under the influence of the electrostatic field of the approaching ion. Massey[31] made a quantum

(31) Massey, H. S. W., Proc. Camb. Phil. Soc., 26, 386, (1930)

mechanical calculation of the neutralization of a positive hydrogen atom when colliding with a metal surface and found the average distance at which the transition takes place to be of the order of angstroms. He found the probability of a transition for the positive ions approached unity at a small distance from the metal surface.

Recent investigations of secondary emission from metals by low velocity positive ions by Hagstrum[32][33][34][35]

(32) Hagstrum, H., Phys. Rev., 104, 672, (1956)

(33) Hagstrum, H., Phys. Rev., 96, 325, (1954)

(34) Hagstrum, H., Phys. Rev., 96, 336, (1954)

(35) Hagstrum, H., Phys. Rev., 89, 244, (1953)

have given information regarding the possibility of an interaction between the approaching ions and the electrons in the phosphor. In his work with metals, results indicate that a process of potential efection rather than one of kinetic ejection predominates in the region of energy less than 1000 electron volts.

Potential ejection is the term used for those cases in which the interaction between the ion outside the lattice and the electrons inside may be approximated by a purely electrostatic field. Ejection can occur when the potential energy recovered on neutralization of the ion is sufficient both to extract the neutralizing electron and to excite a second metal electron to the level above the potential barrier at the surface of the metal.

Electronic transitions in which an excited atom or ion could become involved near a metal surface are resonance, Auger, and radiative types. Massey[36] has shown

(36) Massey, op. cit., p. 389

that neutralization takes place within a few angstrom units of the surface. For instance, when Xenon, the gas used in this work is considered, it is easy to show that a thermal ion has a velosity of approximately 10^4 cm/sec, therefore would spend only about 10^{-12} sec in a neutral state near the surface, making radiation unlikely since the lifetime for radiation (10^{-8} sec) is long compared to this value.

The resonance transition consists of the ion being neutralized to an excited state by an isoenergetic capture of a metal electron. The atom thus formed decays to the ground state with the excitation of a second electron. This process is shown schematically in Fig. 5.[37]

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(37) Hagstrum, H., Phys. Rev., 104, 681, (1956)

In the Auger neutralization process the ion is neutralized directly to the ground state by a metal electron with a second metal electron obtaining the excess energy from the process. This process is shown in Fig. 6.[38]

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(38) Hagstrum, H., ibid., p. 683

Hagstrum was able to explain quantitatively the secondary emission of electrons from tungsten by 10-200 ev He, Ne, A, Xe, and K ions on the basis of the Auger neutralization process before the kinetic processes become important.[39] In fact Hagstrum[40] states that the Auger

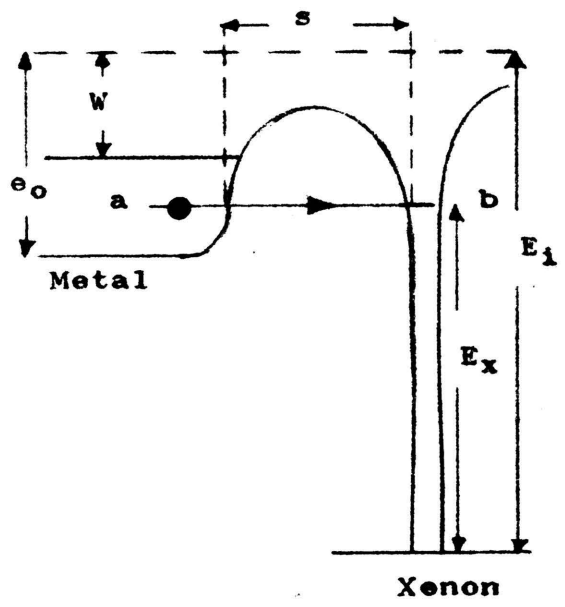


Fig. 5

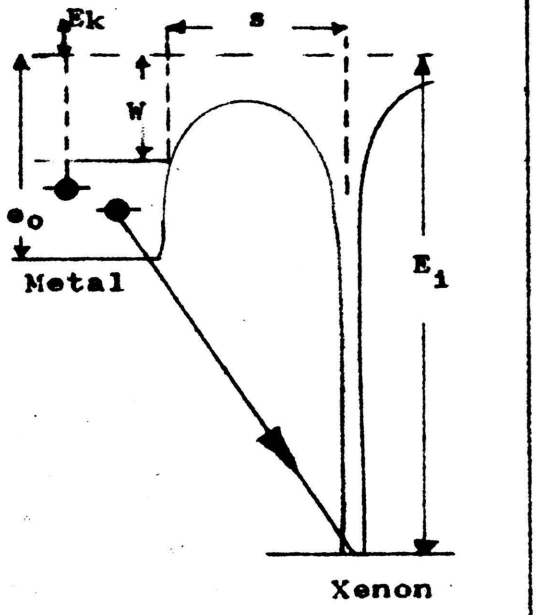


Fig. 6

Fig. 5. Energy level diagram indicating the resonance tunneling transitions which can occur from a metal into singly ionized Xenon. Tunneling into the ion can occur between a and b'as this is the region of a large density of filled levels in the metal and excited levels in the ion.

Fig. 6. Energy level diagram indicating the electronic transitions which occur when the normal singly charged ion of Xenon is neutralized in the process of Auger neutralization.

Definitions of notation used in the figures.

E_i Ionization energy

E_x Excitation energy

s Distance of particle from metal surface

e_o Energy of vacuum level above bottom of conduction band

W Work function of metal

E_k Kinetic energy of particle outside metal

(39) Hagstrum, H., Phys. Rev., 89, 244, (1953)

(40) Hagstrum, H., Phys. Rev., 96, 328, (1954)

processes are so probable that the magnitude of secondary emission in this energy range is determined by the probability that the excited electron will escape rather than by the probability that the process occurs while the ion is near the metal surface.

If we extend this mechanism to the field of non-metals and in particular phosphors we find that the important factors to consider are the ionization energy of the incoming ion and the energy required to extract an electron from the filled band to a region just outside the metal surface and the band gap. This involves a knowledge of the energy width from the top of the filled band to the surface of the phosphor, the width of its filled band and the width of the forbidden gap. An Auger type energy level diagram indicating the important values for semi-conductors is shown in Fig. 7.

According to Leverenz,(41) direct experimental infor-

(41) Leverenz, H. A., Luminescence of Solids, John Wiley and Sons, New York, 1950, p. 119

mation concerning energy level diagrams and the widths of the forbidden gap between the filled and the conduction band is a major lack in phosphor research. In general the

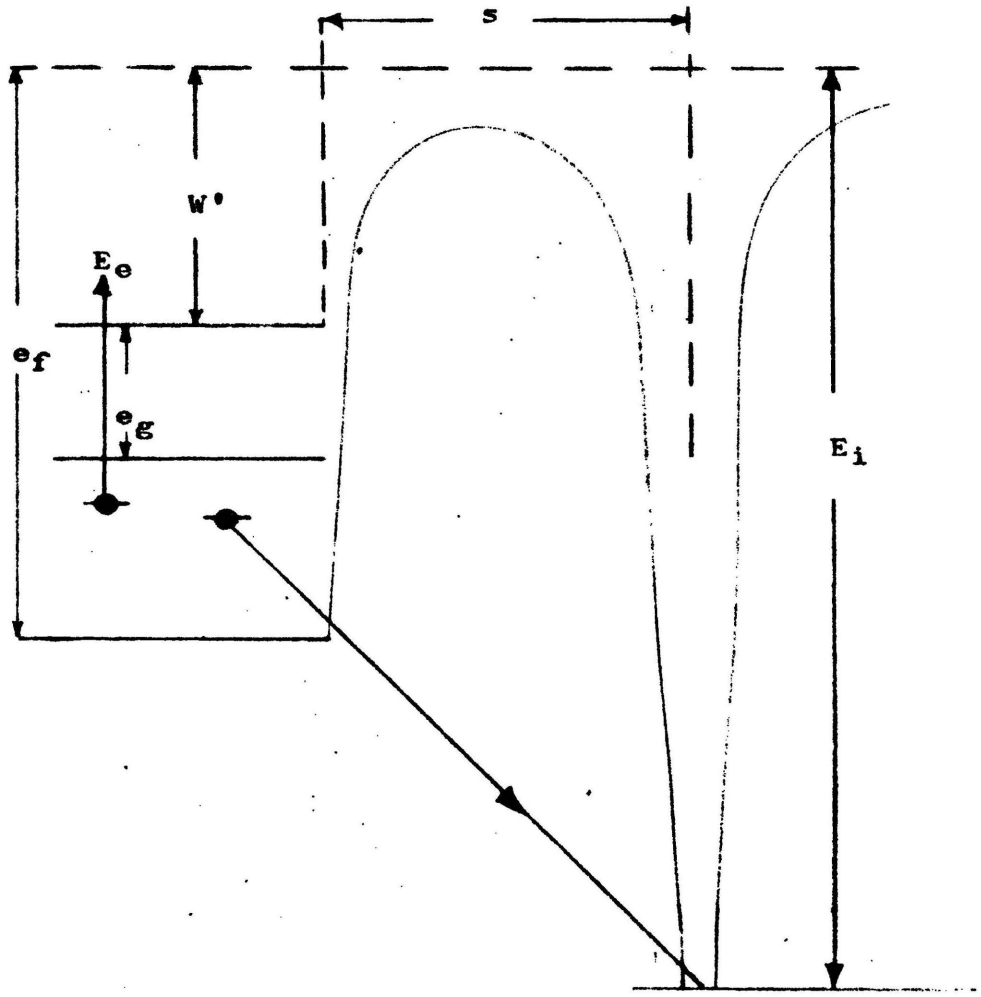


Fig. 7. Proposed energy diagram for Auger type transitions in semi-conductors.

Notation is the same as that in Figs. 5 and 6 with the addition of the following.

E_e Energy given to electron in the filled band

e_f Energy of vacuum level above bottom of filled band

e_g Width of the forbidden gap in electron volts

W' Energy from the top of the conduction band to the surface of the phosphor

width of the forbidden gap in phosphors which have been investigated is about 3 electron volts. A particular lack of information regarding the work functions of phosphors has been noted by the author.

If the Auger process is feasible in the bombardment of phosphors with low energy ions then, referring to Fig. 7, an energy E_e which is the difference between the ionization energy and the energy width from the top of the filled band to the vacuum level will be imparted to electrons in the filled band. If this energy is greater than e_g, the forbidden band gap, electrons will be excited to the conduction band thus triggering the luminescent processes. The ionization energy of Xenon is 12.13 ev (42)

(42) Peaslee, D. C., and Mueller, H., Elements of Atomic Physics, Prentice Hall, New York, 1955

and taking the average forbidden gap for phosphors to be 3 ev we find that phosphors with an energy width from the top of the filled band to the surface of the phosphor of less than 6 ev can have electrons excited from the filled band to the conduction band and trigger the luminescent process. The process could occur with higher values for the work function if there are donor levels present and the Fermi level lies near the top of the forbidden gap.

If the method of kinetic excitation presented by Leverenz is actually the mechanism for low energy ions,

it would seem that it is not feasible to detect low energy ions by scintillation methods. For once the particle has entered the phosphor it seems more probable that the energy would be dissipated in heat than transmitted to the luminescent centers.

Review of Literature

In a review of the literature concerning luminescent materials and detection of charged particles using the scintillation technique, the author was unable to find any articles which dealt specifically with the problem of detecting low energy ions through the use of phosphors.

Leverenz (43) in his comprehensive book on luminescent materials and their uses discusses excitation by charged particles but deals mainly with high energy electrons. This volume does however, furnish information as to which materials might be suitable detectors of low energy ions. Two materials which stand out are Hex ZnO:Zn, which appears to be one of the best detectors of low energy electrons and ZnS which is reported to be a very efficient detector of alpha particles. The zinc oxide phosphor has a very short decay time while the zinc sulphide is a medium persistance phosphor. Both have their primary emission spectra in the middle of the visible spectrum.

(43) Leverenz, H. W., Luminescence of Solids, John Wiley and Sons, New York, 1950

The luminescent efficiency is generally considered to be the ratio of the total light output to the total energy input without questioning which part of this energy is absorbed by the luminescent material, lost by reflection and transmission or dissipated in other processes.

Pringsheim (44) discusses the efficiency of phosphors

(44) Pringsheim, op. cit., p. 29-31

bombarded by charged particles and states the average efficiency is of the order of a few percent, rarely exceeding ten percent for electrons in the kev range. A large part of the primary energy is converted into heat and secondary electron emission. Pringsheim also indicates the existance of a current saturation value for the luminescent yield of phosphors. This usually occurs at currents of five microamperes or less per square centimeter and is dependent upon the energy of the particle beam. The reason for this saturation is that when the available centers are all excited, the luminescence cannot be increased. By increasing the energy of the incident beam, a greater depth of penetration into the phosphor is achieved and consequently more luminescent centers are available for excitation.

For normal screen thicknesses of approximately 10 mg/cm^2 the light emitted from the crystals farthest away from the observer will probably suffer an attenuation of approximately fifty percent in comparison with the light emitted by the crystals nearest the observer.(45)

(45) Nottingham, W. B., Cathode Ray Tube Displays, McGraw-Hill Book Company, New York, 1948, p. 622

Since the phosphor material beyond the luminescence centers acts to reduce the amount of light transmitted to

the photomultiplier tube, a phosphor coating which is quite thin seems to be desirable for use in a detection device for low energy ions.

Zinc Sulphide is one of the few simple inorganic compounds which can be excited to strong luminescence in the crystalline state only, apparently without containing any activating impurity. It is the general belief that the interstitial zinc atoms play the part of an activating impurity in zinc sulphide.[46]

(46) Randall, J. T., Proc. Roy. Soc., London, A170, 272, (1939)

Garlick[47] states that the efficiency of excitation

(47) Garlick, op. cit., p. 200

of zinc sulphide phosphors by alpha particles in the kev range may be as high as eighty percent as compared to the value of ten percent for cathode rays.

Brown[48] discussed the brightness of cathodolumines-

(48) Brown, T. R., J. O. S. A., 27, 180, (1937)

cence in willemite (Zn_2SiO_4:Mn) at low current densities and low voltages. He encounters a dead voltage in the vicinity of 150 volts and indicates that in the region from 150 to 800 volts the brightness is directly proportional to the square of the voltage. The brightness was determined visually by comparison with an equal brightness

photometer which was color matched to the target. This method would indicate that willemite could possibly emit sufficient light under bombardment by low energy particles to be suitable for detection of low energy ions. Leverenz[49]

(49) Leverenz, H. W., Luminescence of Solids, John Wiley and Sons, New York, 1950, p. 238

states willemite will give visible luminescence at energies as low as 12 ev but only when subjected to enormous current densities.

In the course of work on a radio-frequency spectroscope, Kaisel and Clark[50] found it necessary to observe

(50) Kaisel, S. F., and Clark, C. B., J. O. S. A., 44, 134, (1954)

visually patterns created by electrons with energies of the order of five electron volts. A luminescent screen which would react to these energies at low current densities was sucessfully constructed using Hex ZnO:Zn phosphor. The phosphor was settled on glass plates to which had been applied a transparent conducting coating. In this way the incident charge is removed by conduction through the phosphor to a conducting base. In order that the beam current can be removed by conduction intimate contact must be made between the conducting plates and the phosphor particles. Since phosphors are, in general, good insulators the coating must be as thin as possible, yet keeping the density

high for maximum light output. It was found that the addition of binders increased the voltage at which luminescence first was visible and reduced the phosphor sensitivity (i.e. light output for a given incident beam power).

The zinc oxide screens were found to be very susceptible to "poisoning" which occurs from the introduction of undesirable impurities and was discussed earlier in this thesis. Vapors from a diffusion pump, if allowed to enter the region where the luminescent process is taking place, reduced the sensitivity of the phosphor and a dead voltage begins to appear.

In a detailed study of the zinc oxide phosphor by Schrader and Kaisel[51] an apparent violation of Stokes

(51) Schrader, R. E., and Kaisel, S. R., J. O. S. A., 44, 135, (1954)

law was observed in that the luminescence emission of zinc oxide phosphors is a band ranging from 4100 angstroms to 6800 angstroms (3.1 ev-1.8 ev) and it can be excited by electrons with applied voltages as low as 2.4 volts. Hahn[52] has investigated the electrical properties of

(52) Hahn, E. E., J. Appl. Phys., 22, 855, (1951)

zinc oxide and has found a high density of donor levels .04 ev below the conduction band. Avalue of 3.2 ev for the width of the gap between the filled and conduction band has been given by Schrader and Leverenz.[53]

(53) Schrader, R. E., and Leverenz, H. W., J. O. S. A., 37, 939, (1947)

From a review of the literature it seems apparent that zinc oxide is the most promising phosphor for use in detection of low energy particles. It has a short decay time, less than 10^{-6} seconds, and produces visible luminescence under bombardment of electrons with energies of 5 ev and less. It also has the advantage of having been well investigated in the low energy range. It would appear that if low energy ions are able to excite phosphors with a reasonable degree of efficiency that zinc oxide with an abundance of donor levels .04 ev below the conduction band would be likely scintillation material in the field. There also exists the possibility that additional information concerning the luminescence process itself could be obtained working with this phosphor.

The tentative energy level diagram for zinc sulphide as shown in Fig. 8, has been proposed by Bube (54) for the

(54) Bube, R. H., Phys. Rev., 90, 79, (1953)

excitation and emission spectra. The forbidden gap is 3.5 ev wide. Energy levels due to zinc interstitials are found .5 ev below the conduction band and .5 ev above the filled band, it is assumed that these act as luminescent centers in the emission process. There is a high density

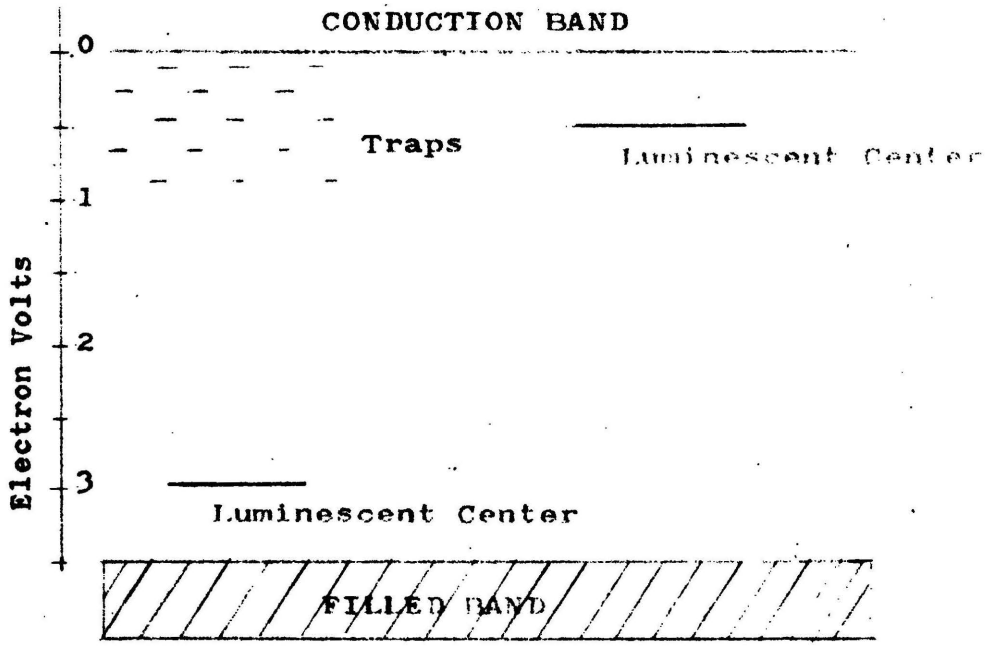


Fig. 8. Proposed Energy Level Diagram for Zinc Sulphide

of traps in the region of 0-1 ev below the conduction band.

Enough information is known about the energy levels of zinc oxide and zinc sulphide to make these phosphors suitable for study in regard to the Auger type transitions which were discussed earlier.

Willemite, due to its demonstrated characteristics under bombardment by electron beams of low current density and low voltages is another likely prospect.

Although it is possible that there are other phosphors, unknown to the author, which would be suitable for the detection of low energy ions, it was decided to use zinc oxide, zinc sulphide and willemite as the scintillation materials in this research.

Samples of the above mentioned phosphors were donated by the Chemical and Physical Laboratories of the Radio Corporation of America. Special thanks are given to Dr. Arthur Smith for his help in obtaining these samples.

Equipment and Experimental Technique

The phosphors to be used are primarily known for their detection of low energy electrons. Consequently, a simple test of the adequacy and purity of the phosphor is to bombard it with electrons. A readily obtainable source of electrons is an electron gun of the type used in conventional cathode ray tubes. Once the existance of an electron beam is ascertained and the phosphor reacts to it satisfactorily, a gas can be introduced into the area and the electron beam can be used to produce ions.

The electron gun comprises those electrodes that create, control and focus the beam of electrons. It consists of a heated cathode as the source of electrons, and one or more electrodes that form and accelerate these electrons into a beam that travels axially down a tube. The beam is controlled in intensity by a negative control grid consisting of a pierced diaphram immediately in front of the cathode. In front of this grid may be an accelerating grid, which is usually at a potential of several hundred volts positive with respect to the cathode. An additional element of the gun may consist of a focusing electron lens, which makes this beam converge to a small area or spot on the front of the tube.

There was no requirement in the detection of ions for deflection of the electron beam so the deflection plates were not needed. The guns used in the first runs of the experiment were donated by Dr. Eugene B. Hensley

of the Physics Department of the University of Missouri. These guns were of the electrostatic focus, low voltage type, modified by removing the deflection plates completely. Due to the fact that the heater filaments for the cathodes of these guns burned out before definite results could be obtained it was necessary to obtain a second set of guns. These were also low voltage, electrostatically focused, and were donated by the Rauland Corporation of Chicago, Illinois.

The production and detection of electrons and positive ions took place in the tube shown in Fig. 9. This tube was designed to provide a distance of approximately 9 inches between the last anode of the electron gun and the phosphor coating. This is the distance used most commonly in cathode ray tubes utilizing this particular type of gun. Side plates were placed almost immediately in front of the last anode of the electron gun. Ions produced by electrons bombarding a gas introduced into the tube can be attracted to the phosphor by applying an electric field between the side plates.

This tube was further modified after a series of initial runs. It was found that the electron beam produced a spot of light easily visible with the eye on the end plate but the side plates were rather inaccessable to the beam. Also the scintillations could be better detected by the photomultiplier tube if it could be placed in direct contact with a large area of the side plates.

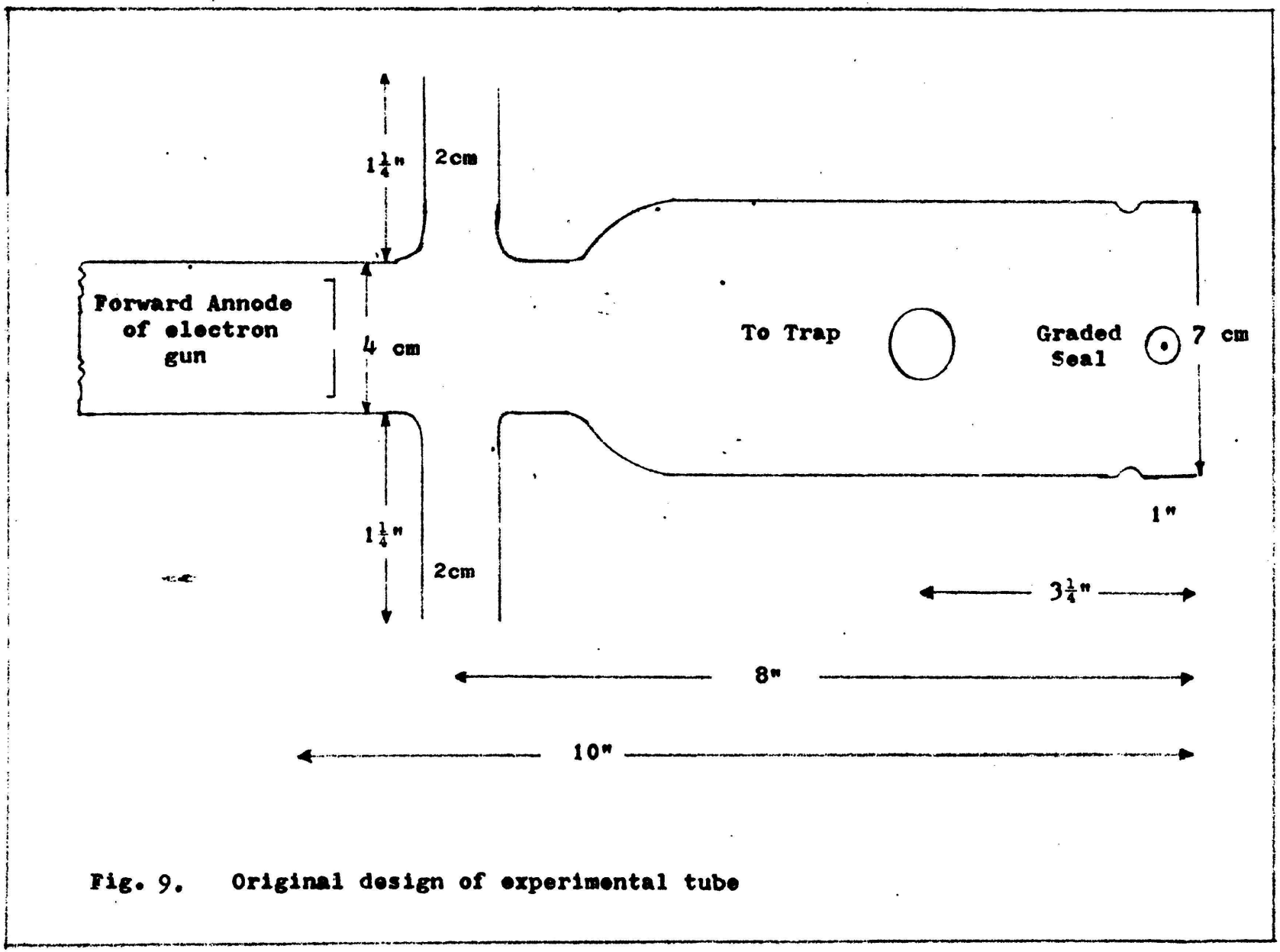
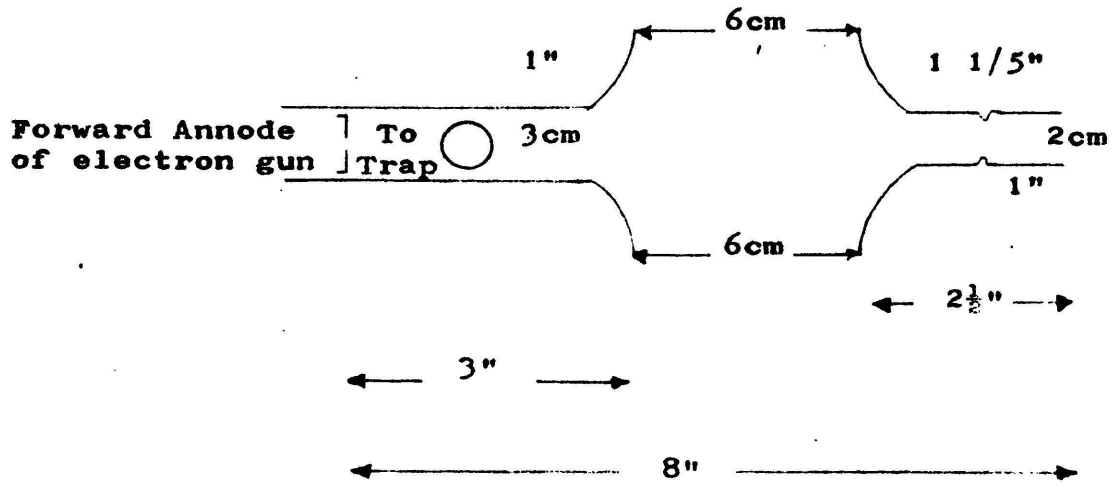
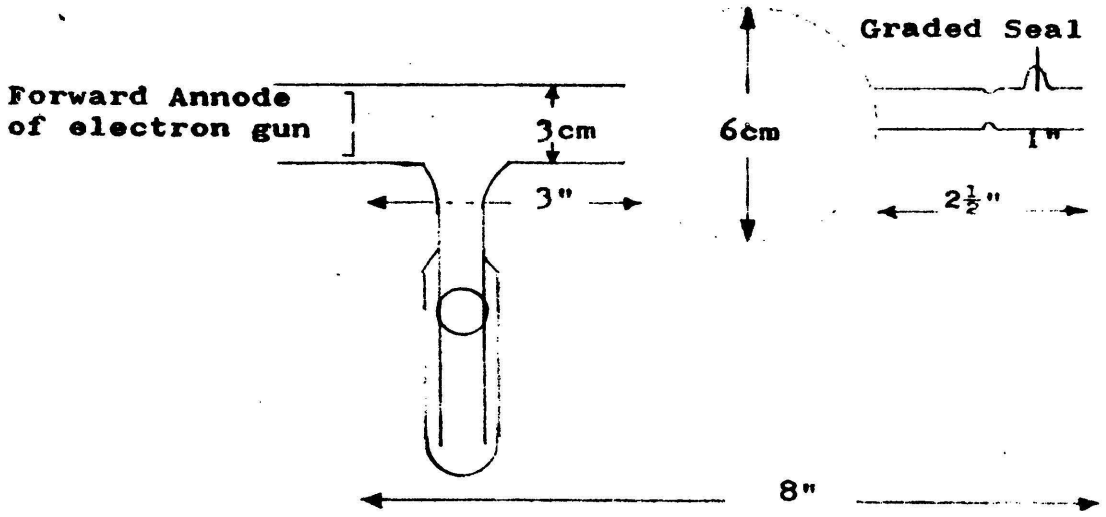


Fig. 9. Original design of experimental tube



a. Top View



b. Side View

Fig. 10. Modified Design of Experimental Tube

Consequently the tube shown in Fig. 10 was built. The main modification was to increase the size of the side plates used to detect positive ions and reduction of the end plate size.

To cut down the light reflected from the gun assembly an interior coating of a matte black material must be used. A number of substances are suitable for this purpose, e.g. Acquadag, Acquagraph, Dixonac, and carbon black in sodium silicate.(55) A coating of sodium silicate and carbon

(55) Zworykin, V. K., and Merton, G. A., Television, John Wiley and Sons, New York, 1954, p. 76

was tried but if was found to trap and retain considerable quantities of water vapor even after baking at 200° C. for six hours. A satisfactory coating was obtained by mixing carbon black with high vacuum wax (Apiezon W) dissolved in carbon tetrachloride. This proved to have a low vapor pressure and provide a consistent light absorbing coating.

The electron gun was provided with external connections through a five-lead press which was connected to the gun leads. This enabled control over the filament current, cathode voltage, control grid, focusing anode, and accelerating anode. A schematic of the gun and external connections is shown in Fig. 11.

The entire system was made of glass. This reduced outgassing problems to a minimum and facilitated leak

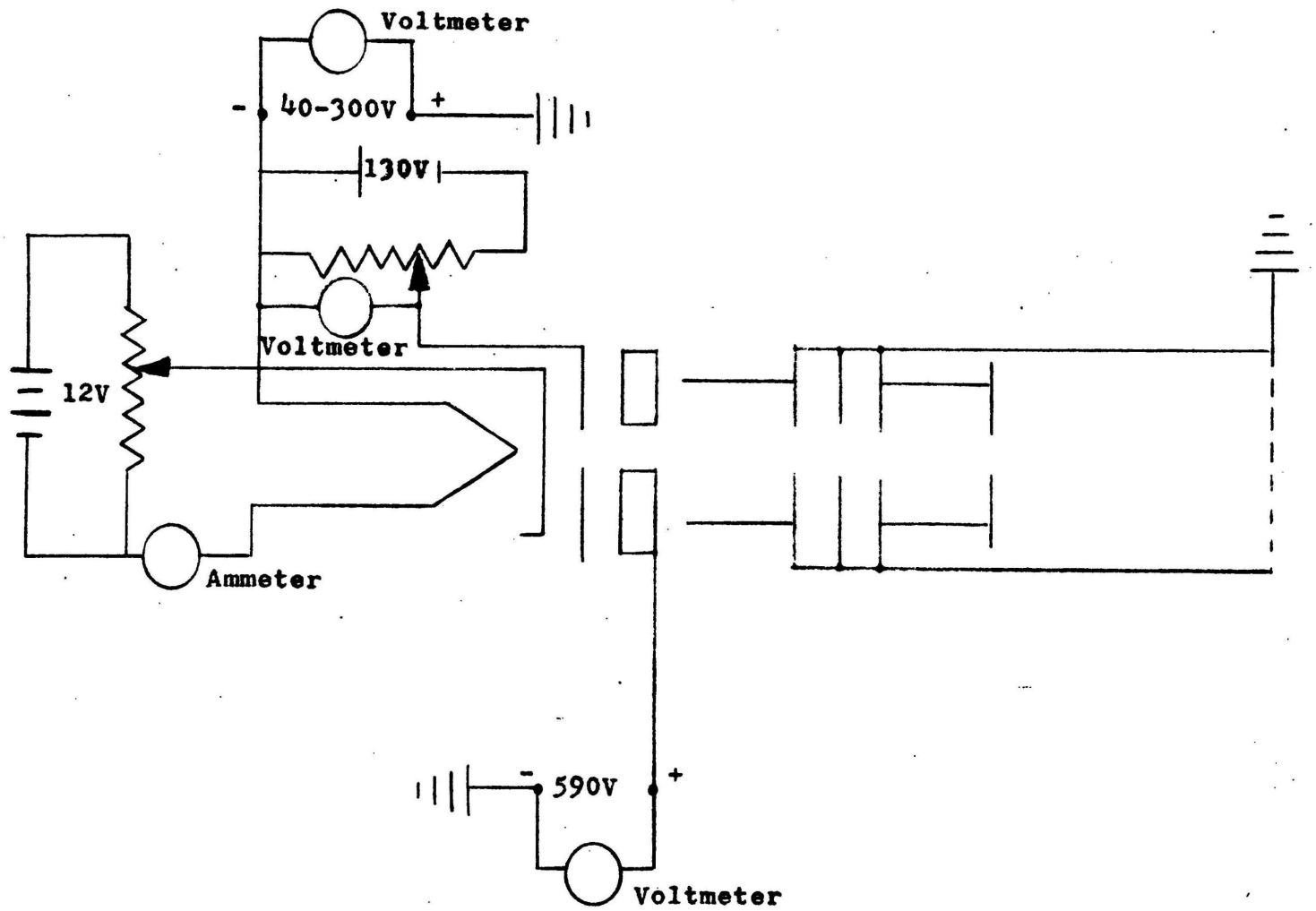


Fig. 11. Schematic of Electron Gun and External Connections

detection. The pumping apparatus consisted of a Cenco HyVac forepump and a two stage umbrella type mercury diffusion pump. The pressures attained were read by a McLeod Guage connected into the system. A series of stopcocks located throughout the system enabled one to close off parts of the system, and to leak air or gas into the system. A trap, cooled by an acetone and dry ice mixture, was placed between the pumps and the experimental tube to prevent the contamination of the phosphors by oil or mercury vapors. A block diagram of the vacuum system is shown in Fig. 12.

To reduce a variation of the energies of the electrons while testing the phosphors and the beam the pressure was reduced to 10^{-5} millimeters of mercury or less. At this pressure the mean free path of the electrons was 5 meters or greater. Consequently the probability of collisions by the electrons before reaching the phosphors was small.

Xenon gas was introduced into the chamber from the resevoir shown in Fig. 12 through a series of stopcocks. Xenon was used as a source of positive ions because it has few metastable states and the noble gases form negative ions by attachment much less readily than most other gases. The presence of metastable states is undesirable for the average life is long (10^{-3} sec or more) and at any time may produce ionization of the gas or by photon release may in-

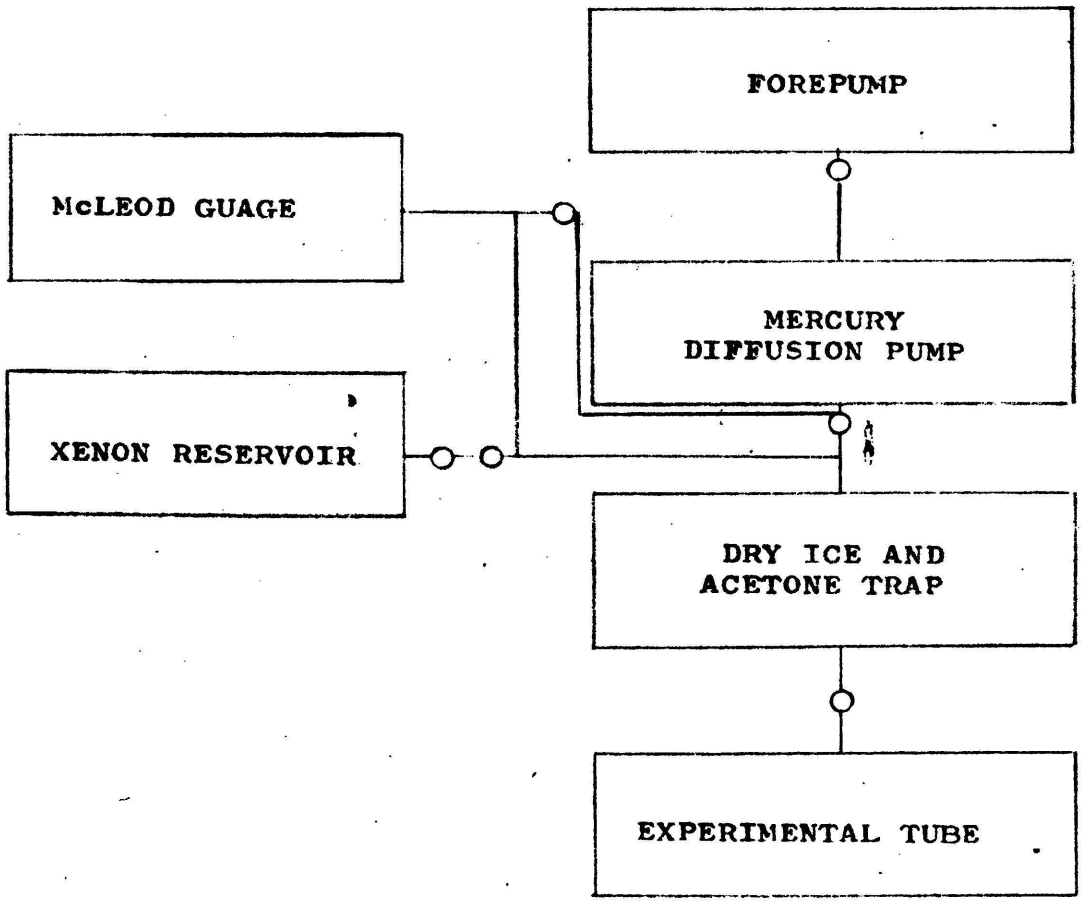


Fig. 12. Block Diagram of Vacuum System

troduce spurious light. An additional reason for using the noble gases was so that there would be no question as to the type of ion present such as N_2^+ or N^+. Xenon is also massive, which allows one to have a reasonable time interval between a pulse and an ion striking phosphor.

The phosphors were coated on special conducting glass plates known as E. C. conducting plates, manufactured commercially by the Corning Glass Works. The base of the plate is standard Pyrex brand glass. To the surface of this plate is permanently bonded a thin, transparent, electrically conductive film. This film is quite highly transparent with light transmission up to 70 percent. It is also chemically inert and extremely hard. The coating can be cleaned by normal cleansers but due to its extreme thinness, about 10^{-6} inches, precautions must be exercised to avoid scratching the coating with harsh abrasives. It was found by the author that carbon tetrachloride was an effective cleanser.

The maximum recommended operating temperature for these plates is 350° C. and contact with flame should be avoided. It was found during the experimental work that the coating was damaged if a gas discharge was allowed to begin in the tube. Electrical connections can be made to the conducting plates by copper strips which utilize a battery clamp to provide a pressure contact with the conducting coating.

These plates were sealed to the arms of the main tube

with high vacuum wax.

Since low energy charged particles are affected by the earths magnetic field it was necessary to determine its flux density and dip angle and construct a device to neutralize it. The field was determined to have a horizontal component of .270 gauss and a vertical component of .521 gauss. The angle of dip was found to be 62° 40' at our location. These results were obtained through the use of a Cenco Earth Inductor Coil and a ballistic galvanometer. The field was canceled by producing an equal and opposite field with a rectangular cage which enclosed the experimental tube. A current of .91 amperes in the windings reduced the field to a value of one half percent or less of its original value in a plane 47 centimeters above the table top.

The phosphor was placed upon the Plates by a method of gravity settling. Gravity settling usually produces a uniform screen with the larger particles in direct contact with the glass and the smaller ones filling in the interstices between the larger particles. Methyl alcohol was chosen as the liquid in which to suspend the phosphors in preference to distilled water due to the greater ease in producing a solution in alcohol and the rapid drying of the plates. A detailed outline of the plating procedure is included in Appendix C.

Since the thickness of the coating can be quite critical it is desired to have as thin a coating as possible

but yet to have complete coverage. It was finally decided to use a coating of one half milligram per square centimeter. A satisfactory empirical rule for phosphor coatings seems to be that they should be thin enough to allow a shadow of the cathode to be visible on the reverse side. According to Nottingham (56) 1000 volt electrons will penetrate phosphors about .4 micron. Since the thickness of the screen is determined by the distance of penetration of particles and the average size of the phosphor particles being used is three microns it seems that the most desirable screen would be one particle layer thick.

(56) Nottingham, W. B., Cathode Ray Tube Displays, McGraw-Hill, New York, 1948, p. 609

Since the particle size of the phosphors we are using runs about three microns we are close to the realm of pseudocolloids. We must therefore observe some of the principles of colloidal phenomena in our settling suspensions. One empirical law (Coehn 1898) of colloid physics is: If a colloidal dispersion consists of two dielectrics, the substance having the greater dielectric constant charges itself positively with respect to the substance of lower dielectric constant. (Note: Alcohol e=56.6; ZnO e=9)

In settling of the zinc oxide screens trouble was encountered in that the screens looked like force lines traced by a myriad of tiny iron filings in a magnetic field. Vigorous shaking failed to eliminate these stream-

er-like volumes, approximately 1/16 to 1/4 inch wide, which were visibly distinguished by having very few suspended particles.

This problem can be solved by introducing into the suspension a soluble substance which has a high dissociation thus procucing mobile positive ions to discharge the negatively charged phosphor particles as fast as they are formed on falling through the solution. This was achieved by the addition of a solution of dilute Hydrochloric acid giving mobile positive ions.

The scintillations obtained by the charged particles in most cases can be observed visually. But the apparatus was designed so that a photomultiplier tube could be used as a detector device. The tube used was a DuMont 6292 Photomultiplier tube. This is a ten stage phototube of the end window type thus facilitating placing the phototube in direct contact with the phosphor plate. The current amplification of the tube is 10^5 with dynode voltages of 105 volts per stage. The photocathode is most sensitive to the spectral region between 4300 and 5300 angstroms which matches well with the emission spectra of the phosphors. This is one criterion for peak counter performance of the tube. The dark current of this tube with an applied voltage of 1000 volts is .0736 microampere at room temperature and by cooling the tube with a mixture of dry ice and acetone (-76° C) this can be re-

duced to .0064 microampere.

The output of the photomultiplier tube can be read directly to obtain the magnitude of the direct current from the tube. This would necessitate insuring that ambient light from exterior sources or from the tube filament be shielded in order that the results be indicative of the scintillations produced. Even if such shielding were possible the further question arises as to the detection of only positive ions. There exists the probability of electrons and photons also striking the phosphor and producing scintillations.

To be reasonably certain of the results the following procedure was used. A field was applied across the two side plates making one positive and the other negative. Therefore the ions and the electrons would go to opposite sides. Furthermore it was calculated that the time required for a one hundred volt potential difference to sweep these ions from the axis of the tube to the plates is of the order of four microseconds.

By pulsing the electron beam with microsecond pulses at a reasonable frequency, pulses of ions should be produced. These pulses of ions should produce scintillations a given time after the electron pulse if the phosphor will detect. Due to the difference in velocity of photons, electrons, and ions it should be possible to discriminate between scintillations produced by these particles.

A block diagram of the set-up used is shown in Fig. 13. Pulses with variable length, frequency and amplitude were produced by a General Radio Unit Pulser, Model 1217A. Pulse duration is variable from one microsecond to one hundredth of a second, frequency from 10 pulses per second to 50,000 pulses per second and the amplitude varies from 0-21 volts. These pulses were fed into a linear amplifier so as to obtain output pulses of 100 volts from the amplifier. The positive pulses were then fed into the grid of the electron gun which has a variable bias of 0-130 volts. The photomultiplier tube is placed in contact with the phosphor plate and the output of the tube is directed across a one megohm resistor to a Tetronic oscilliscope which was adjusted to read alternating current signals only. Furthermore the scope was triggered from the amplifier to start a sweep when a pulse was delivered to the grid of the electron gun. The circuit diagrams of the amplifier, power supply for the photomultiplier, and the photomultiplier are shown in Appendix E.

In order to keep unwanted low frequency output from the photomultiplier tube from reaching the oscilliscope a high pass filter as shown in Appendix F was placed in the circuit at the output of the photomultiplier tube.

Precautions to reduce pick-up of the signal by the oscilliscope from the leads consisted in using coaxial cable for all connections transmitting the pulses. The

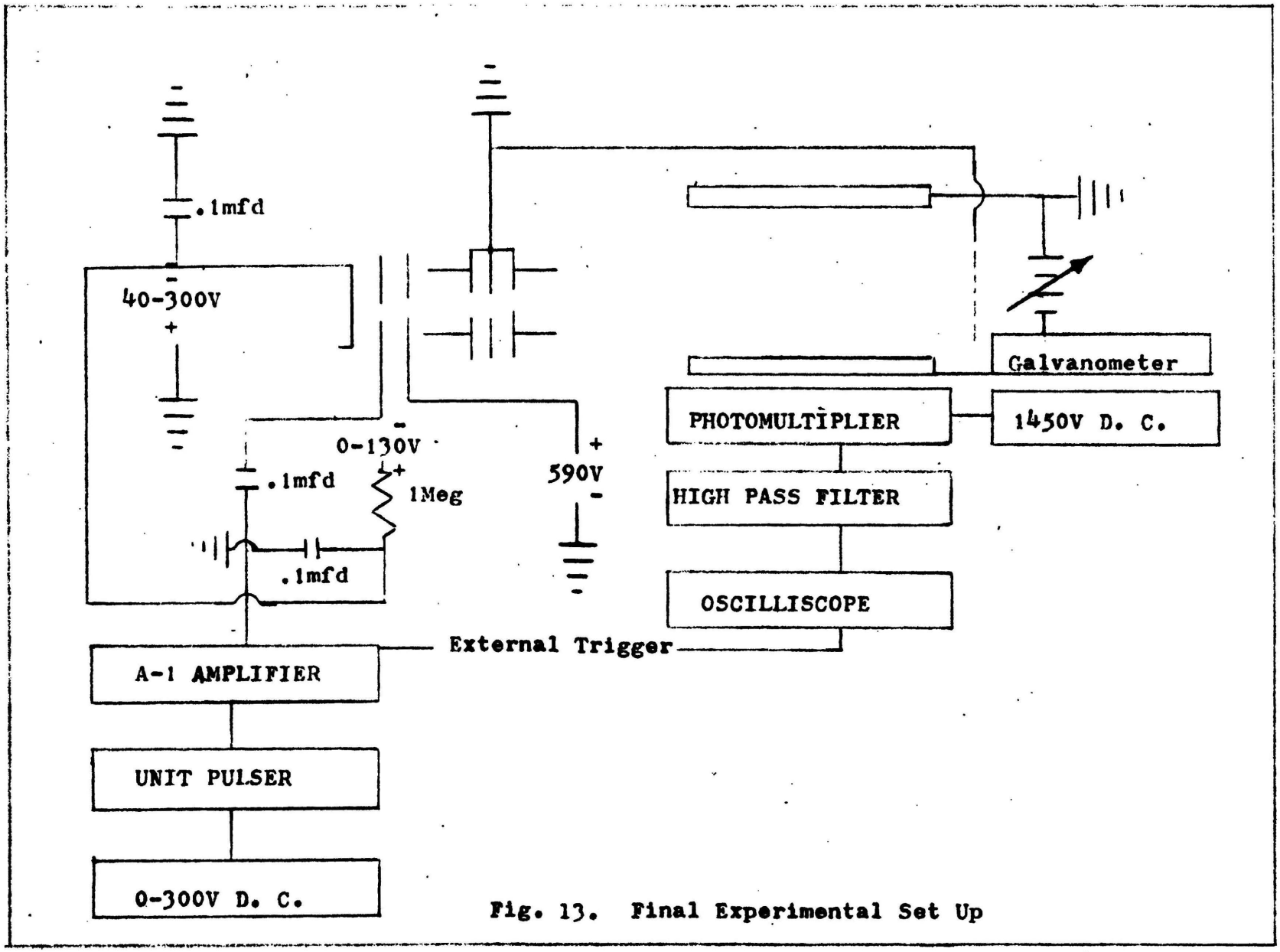


Fig. 13. Final Experimental Set Up

leads from the photomultiplier tube to the base and to the scope were also shielded to eliminate pick-up or transmission of low frequency signals which may be due to filament light or background light.

Using this set-up it was attempted to detect the production of scintillation by ions. The first step in the procedure was to ascertain the existance of a beam of electrons and try magnetically and electrostatically to obtain the best focused beam possible. Under high vacuum the beam was deflected to the phosphor plates and the grid bias used to vary the intensity of scintillations. Xenon gas at a low pressure was admitted to the tube and the gun voltages carefully regulated to prevent a gas discharge from beginning. The beam became very scattered after leaving the last anode of the gun.

After the condition of the beam and the phosphor were checked the photomultiplier was placed in contact with the phosphor plate and pulses applied to the grid of the electron gun. The input pulse could be monitored by changing inputs on the oscilliscope and feeding a signal from a second output of the amplifier to the oscilliscope coincident with the pulse to the grid. The output of the photomultiplier tube went to the high pass filter where the direct current component could be passed through a microammeter to ground and the alternating current component was sent to the oscilliscope which was triggered to

begin a sweep at the time a signal left the amplifier. The current to the phosphor plates was measured by a galvanometer with a current sensitivity of 9×10^{-10} amperes per scale division.

Results and Conclusions

The first set of guns and tubes yielded a focused beam that produced a well-defined spot on the phosphor screen. The scintillations were found to be voltage dependent and the beam could be cut off with a grid bias of - 15 volts.

It was determined that the phosphors were sensitive to low energy electron bombardment at currents of the order of microamperes. When the Xenon was introduced into the experimental tube scintillations were observed visually but it was impossible to determine if these were due to ions, electrons, or photons before the filaments on these guns burned out. It is felt that if the pulse technique could have been applied to these guns, full use of this technique could have been achieved due to the well-focused beam obtained at low voltages.

The electron beam obtained from the second type of gun was not as well focused as that of the first type; it became very diffuse after leaving the last anode. It is possible that higher voltages should be used on the accelerating anode to produce better focusing and also that the allignment of the gun was disturbed when it was mounted. A considerable grid bias was needed (-150V) to cut the beam off. Although the external magnetic field was very effective in deflecting the beam from the first gun it was not nearly as effective on the beam of the second gun. By

using magnets near the tube the beam could be deflected to strike either side plate where it appeared very poorly focused.

The signal obtained through the amplifier from the pulser is shown in Fig. 14. As can be seen the pulse has a sharp rise with a rather long tail. The length of the pulse is of the order of microseconds and the repitition rate is 5000 per second. This should allow a sufficient time between the pulses for the ions to be swept to the phosphor plates. The pulses going into the grid of the electron gun were 100 volt pulses of the same shape as the one shown in Fig. 14.

Shielding reduced pick-up to a value of about one thousandth of a volt. But with the photomultiplier operating at 240 volts for the first stage and 120 volts per stage thereafter, a considerable amount of noise appeared on the oscilliscope. This noise definitely appeared to come from the photomultiplier tube itself and not from the power supply or leads.

While investigating the signal produced, the pressure, grid bias, pulse length, pulse frequency, pulse amplitude, and the voltage between the phosphor plates were varied independently. The photomultiplier current and the current to the phosphor plates could be read at any time. The input voltage sensitivity and the sweep time of the oscilliscope were varied to correspond to the signal received.

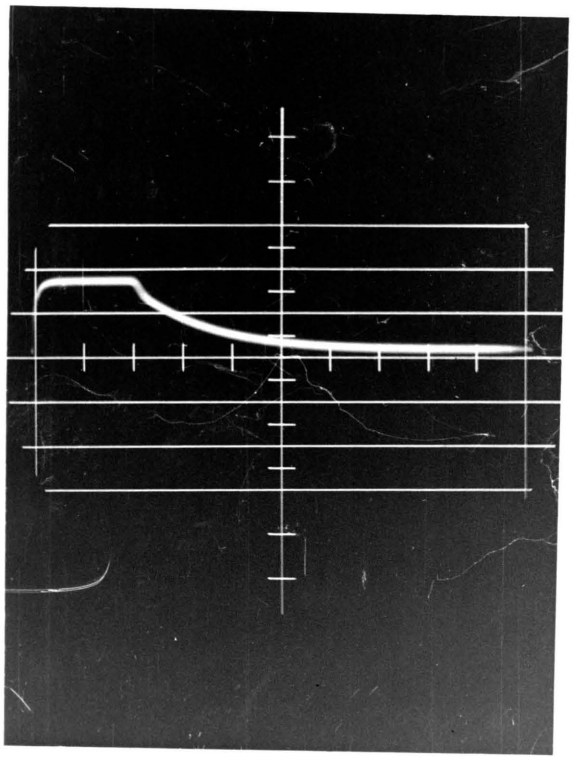


Fig. 14. Oscilliscope trace of the pulse as obtained from the amplifier. The oscilliscope sweep time is 1 microsecond per cm. and the pulse frequency is 5000 per second.

The filament current was kept at .75 amperes during the work and the cathode voltage at -280 volts and the accelerating anode at +590 volts. The variables measured are shown on the sample data sheet in Appendix G.

Considerable pressure fluctuations occurred in the data taking which seemed to be due to outgassing in the main tube. It is not certain whether this was due to the gun or the light absorbing coating on the tube, as the tube itself became quite warm when run for any length of time. Consequently it was difficult to keep a constant pressure for any length of time. This was compensated for by adjusting the pressure to the desired value by pumping on the system before each run and by taking pressure readings before and after each run. Tests with the Tesla coil indicated that the gas in the tube at all pressures was Xenon. The gas never appeared to be air.

At no time during the runs with the second tubes and guns was any current reading obtained from the phosphor plates. This would indicate the effective current was less than 10^{-7} ampere although it is likely the actual current was larger but that part of it is neutralized by secondary electron emission.

The photographs in Figures 15, 16, and 17, show the results obtained from the zinc oxide phosphors with Xenon in the tube. The length of the pulse put on the grid is one microsecond at a frequency of 1000 pulses per second.

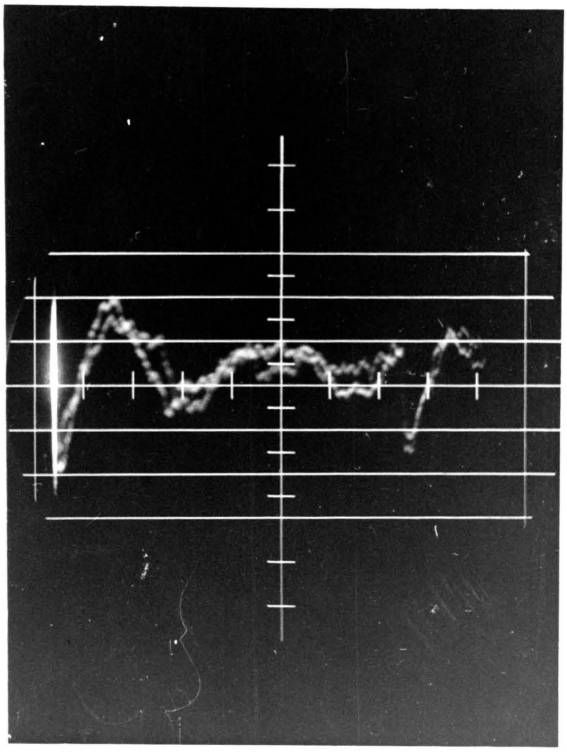


Fig. 15. Signal trace showing results from the zinc oxide with the phosphor plate at ground potential.

Pressure	1.5 micron
Grid Bias	-30 volts
Phosphor Plate Voltage	0 volts
Pulse Length	1 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Sensitivity	.1 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	8 microampere
Phosphor Plate Current	None

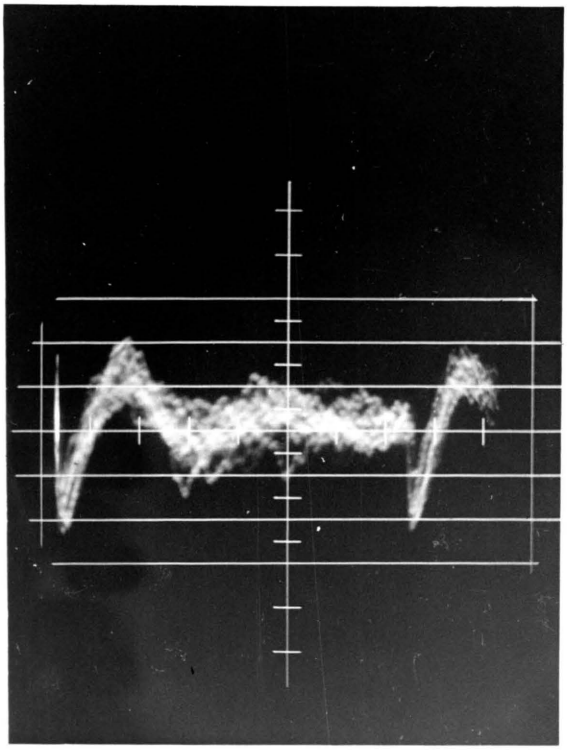


Fig. 16. Signal trace showing results from the zinc oxide with a voltage of -70 volts applied to the phosphor plate.

Pressure	1.9 micron
Grid Bias	-30 volts
Phosphor Plate Voltage	-70 volts
Pulse Length	1 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Sensitivity	.1 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	7 microampere
Phosphor Plate Current	None

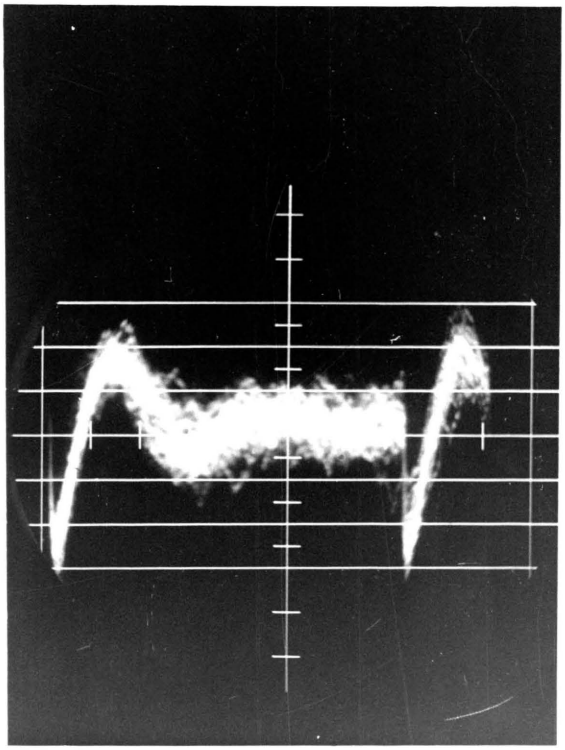


Fig. 17. Signal trace showing results from the zinc oxide with a +70 volts applied to the phosphor plates.

Pressure	1.9 micron
Grid Bias	-30 volts
Phosphor Plate Voltage	+70 volts
Pulse Length	1 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Sensitivity	.1 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	11.8 microampere
Phosphor Plate Current	None

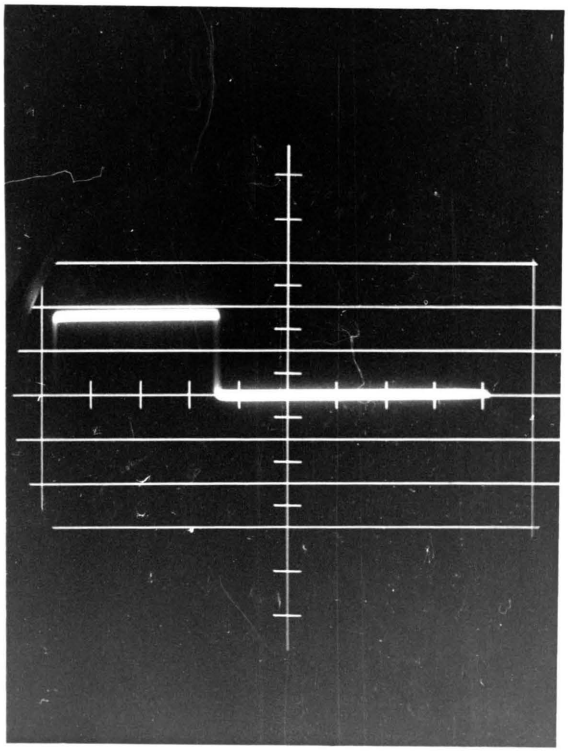


Fig. 18. Calibration voltage signal measuring .15 volt for the signals shown in Figures 15, 16, and 17.

The pressure is about one micron and the grid bias is -30 volts. The variable in these signals is the voltage applied to the phosphor plates. In the first figure the plate is at ground potential, in the second at -70 volts and in the third at +70 volts. The size of this signal can be seen by comparison with Fig. 18, which is a calibration voltage signal of .15 volts. It is apparent that the voltages applied to the phosphor plates have an effect upon the size of the signal and that electrons are being detected. The size of the signal is also affected by grid bias and pressure. Additional photographs of signals produced by changing other variables are shown in Appendix G. It is difficult to explain the shape of the signal but it is possibly due to a gas discharge which starts when the positive pulse is applied to the grid.

The willemite phosphor plates failed to respond to the pulsing technique satisfactorily, probably due to the long time constant (10^{-2} - 10^{-3} seconds) of the willemite. The zinc sulphide phosphor was located in such a position that is was inaccessable to the photomultiplier tube and background light precluded observing visually any change in the emission from the phosphor.

No definite indications as to the detection of positive ions was obtained but it is obvious that if the ion pulses produced output signals of the order of hundredths

of volts these would probably be masked by the noise level alone without considering other competing processes. In addition the shape of the signal produced a sharp downward curve in the region where the expected ion signal would appear, making detection possibilities poor.

It is felt by the author that the technique used is valid and with modifications it would be feasible to discern possible small ion signals. If possible, gas discharge phenomena should be eliminated from the signal. It may be that a solution would be to alter the method of producing the ions. A range of definite energy values for the ion beam would also be desirable. Noise levels may be reduced by using a carefully chosen photomultiplier tube and cooling it to liquid air temperatures. Solutions to the aforementioned experimental difficulties would enhance the possibilities of sucess in further work on this problem.

The effects of electron scintillations were observed, consequently it can be stated that if positive ion scintillations exist they must be of a smaller magnitude than those due to electrons, as expected. The results do not indicate that scintillations due to low energy positive ions exist, nor do they indicate that they don't exist.

Summary

A technique was developed which should be capable of detecting scintillations produced by low energy positive ions. It was found that the method of application of this technique did not possess the resolution necessary to ascertain the feasibility of scintillation detection of low energy positive ions.

The mechanisms involved in the interactions of low energy particles with insulating materials were investigated. The Auger process was projected as a mechanism to justify the feasibility of detection. If detection is possible and an Auger type mechanism is responsible, it should be possible to meliorate the lack of information regarding energy levels in phosphors by applications of the principles involved in this process.

APPENDICES

Appendix A

Strata of the Upper Atmosphere

The ionosphere, as indicated in Fig. A-1,[57] is characterized by the presence of a number of stratified layers of intense ionization denoted in the figure by the letters D, E, and F. Although all parts of the ionosphere contain ionized gases, the intensity of ionization varies with altitude, passing through a number of maxima and minima or layers.

(57) Newell, H. E., High Altitude Rocket Research, Academic Press, New York, 1953, p. 12

The lowest of these layers of maximum ionization is known as the D layer. This layer forms during the daytime at an altitude of approximately sixty kilometers and disappears at sunset. Of the several processes suggested for the formation of the D layer, the following appear to have the strongest support.

$$O_2 + h\nu\ (900\text{-}1100\ A^\circ) \text{ ---------- } O_2^+ + e \quad (58)$$

$$NO + h\nu\ (1100\text{-}1300\ A^\circ) \text{ --------- } NO^+ + e \quad (59)$$

(58) Mitra, S. K., Bhar, J., and Ghosh, G., Indian Journal of Physics, 12, 455, (1939)

(59) Bates, D. R., and Seaton, M. J., Proc. Roy. Soc., London, B63, 129, (1950)

The D layer is an absorbing layer, as far as radio is concerned hence the daytime decrease in signal.

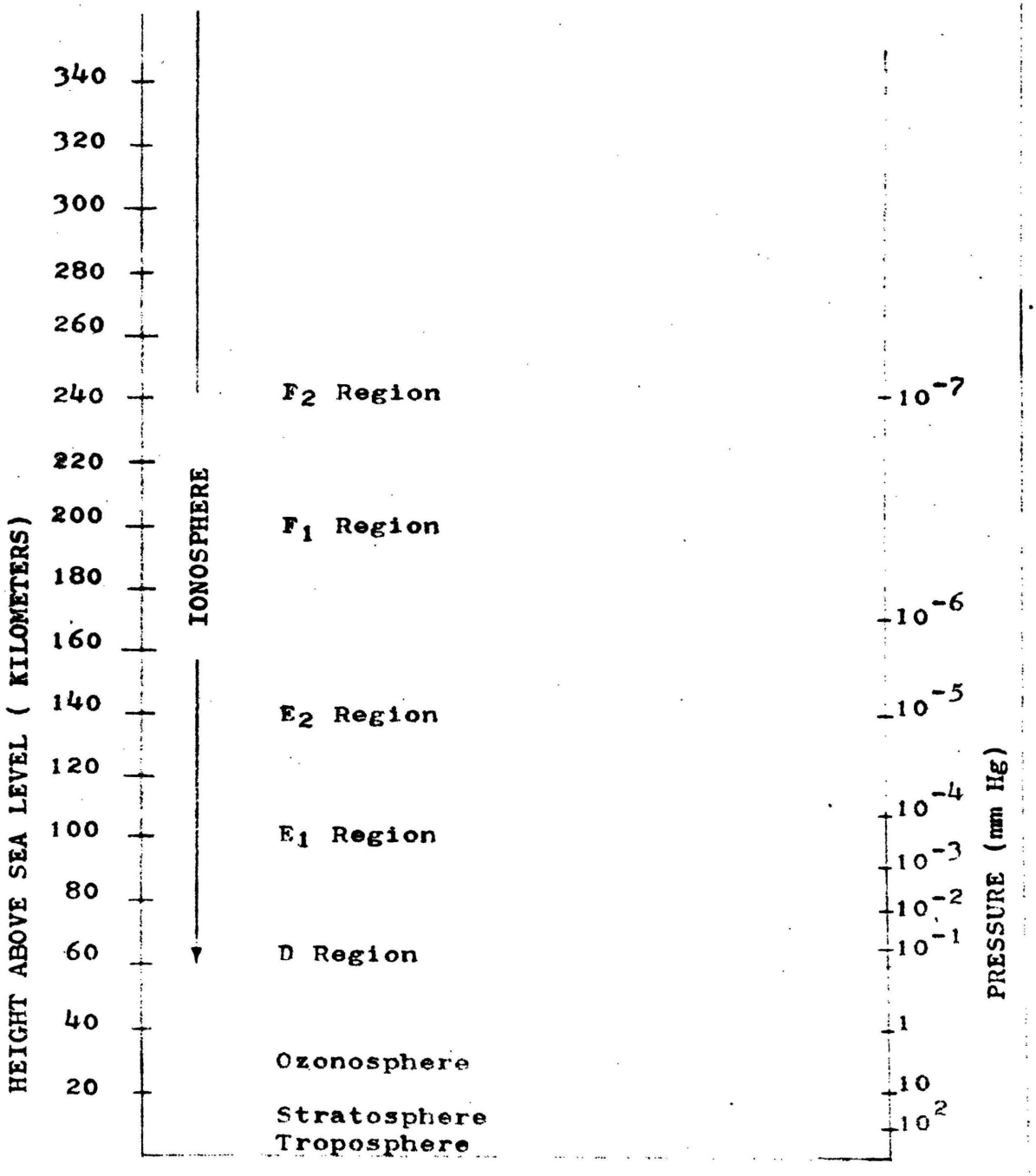


Fig. A-1. Schematic Diagram of the Upper Atmosphere

Above this at altitudes of approximately 100 and 140 kilometers, a pair of more persistent layers known as the E layers are formed. These layers, also known by the name Kennely-Heaviside layers, serve to reflect radio signals back to earth and are of prime interest to the radio-physicist.

Still higher in the upper atmosphere are found another pair of layers known as the F or Appleton layers varying in altitude from 200 to 240 kilometers. Like the E layers these layers are permanent and highly reflective to radio signals. Actually this portion of the ionosphere is composed of two separate layers, the F-1 and F-2, during the daylight hours. At night they coalesce into one F layer. These layers are again important in the transmission of radio signals and have been the object of investigation for many years.

The existance of further layers is speculative. The upper limit of direct measurements, through the use of rockets, have extended to altitudes of only slightly more than 200 kilometers.

Appendix B

The Chapman Theory

Mitra, S. K., The Upper Atmosphere, The Asiatic Society, Calcutta, India, 1952, p. 280

A monochromatic beam of unit cross section is considered as entering the atmosphere at an angle q with the vertical. Let the absorption coefficient per mass of the gas be A and the intensity of the incident radiation be I at height H above the ground. The decrease of intensity after passing through a layer of thickness dh at the height h will be given by:

$$dI = AI\ dh\ \sec q\ p_o\ \exp(-h/H)$$

where p_o is the density of the gas at sea level and H is the "scale height", (H=kT/mg).

Integration gives

$$I = I_o\ \exp(-Ap_oH\sec q\ \exp(-h/H))$$

where I_o is the intensity of the radiation before it enters the atmosphere.

If B is the number of ions produced in the absorption of unit quantity of radiation, the rate of ion production per cubic centimeter at height, h is

$$Q = B\ dI/dh\ \cos q$$

$$Q = B\ A\ I_o\ p_o\ \exp(-h/H - A\ p_o\ H\ \sec q\ \exp(-h/H)).$$

Differentiation of this expression gives Q as a maximum at altitude h given by: $\exp h/H = A\ p_o\ H\ \sec q$ and the maximum rate of ion production will be: $Q = B\ I_o\ \cos q/\ H\ e$.

Appendix C

Plating of Phosphors on Conducting Glass Plates

This method is based on techniques used by Meier Sadowsky as outlined in Transactions of the Electrochemical Society, 95, 112, (1949)

1. Clean all glassware thoroughly. Nitric acid followed by distilled water rinses.
2. Set glass plate in a deep container which in turn is set on a cork ring or other surface free of vibration.
3. Measure out a quantity of the phosphor which will give a uniform coating of the thickness desired.
4. Place phosphor in a mortar and mill gently until there are no sizable lumps evident in the phosphor. Care must be taken not to mill the phosphor too strenuously for it is possible to alter the characteristics of the phosphor in this manner.
5. Place phosphor in a suspension in methyl alcohol which has been filtered to remove any foreign bodies which might be present.
6. Pour solution into deep container and allow to set in a dust free area until the phosphor has settled. Alcohol will become clear upon settling (3-5 hours) and the Tyndall effect can be used to determine when settling is completed.
7. The liquid may be removed by slow siphoning, using a tube of one millimeter diameter or less, well away from the plate. It will be observed that the process

is gradual enough that no disturbance of the coating on the plate takes place.

8. Final drying can be done by air evaporation or by heating with a lamp.

Final result is observed to be a uniform coating. Care must be exercised in handling the coated plates for adherence is not too good. A more durable coating can be obtained by the addition of binders but indications are that these tend to increase the dead voltage and reduce the efficiency of the phosphor.

Appendix D

Characteristics of Phosphors Used in This Research

The information contained in this appendix was obtained from Leverenz, H. W., Luminescence of Solids, John Wiley and Sons, New York, 1950; and information from the Radio Corporation of America.

Basic Material	ZnS	Zn_2SiO_4	ZnO
Activator	Pure	Mn	Zn
Color of Emission	Light Blue	Green	Blue Green
Peak of Emission	$4650A^{o}$	$5250A^{o}$	$5500A^{o}$
Excitation Band	Near UV	Below $2960A^{o}$	Near UV
Duration of Lum.	10^{-5}sec	10^{-2}-10^{-3}sec	10^{-6}-10^{-7}sec
Average Crystal Size	----	3 micron	3 micron
Crystal Structure	Cubic	Rbhdl.	Hex.

Appendix E

Electronics

The schematics shown on the following pages comprise the photomultiplier circuit used with the DuMont 6292 photomultiplier tube, the amplifier circuit used to amplify the pulse output, and the power supply for the photomultiplier tube. The amplifier circuit shows only the portion of the A1 amplifier that was actually used, as the output from the entire amplifier was considerably larger than that needed in the work.

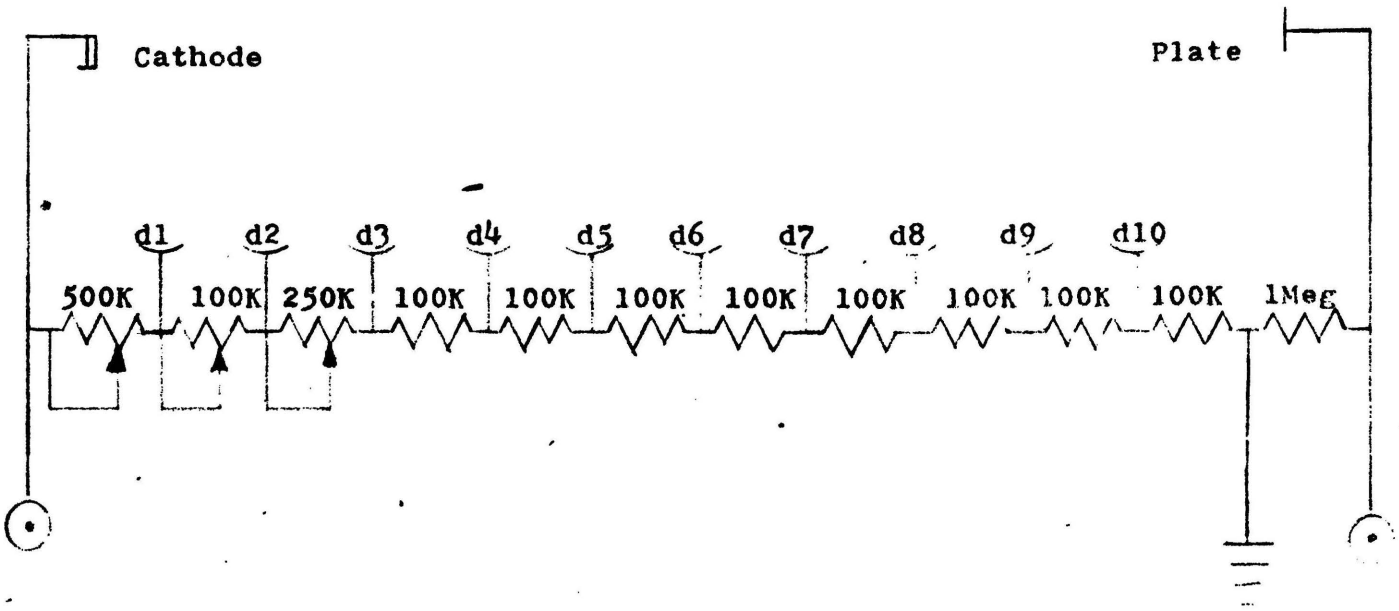


Fig. E-1.. Photomultiplier Assembly

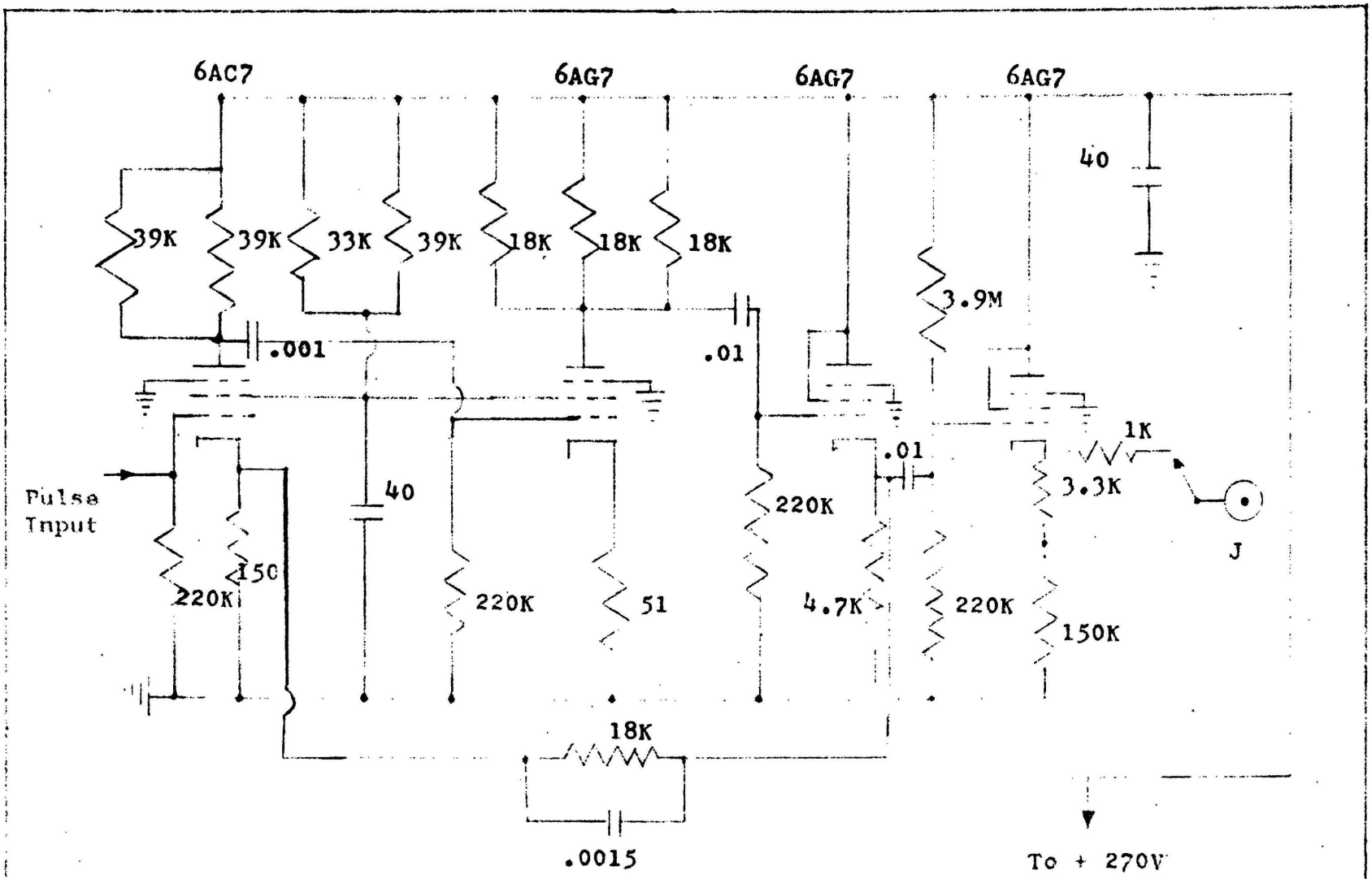


Fig. E-2. A1 Linear Amplifier Schematic

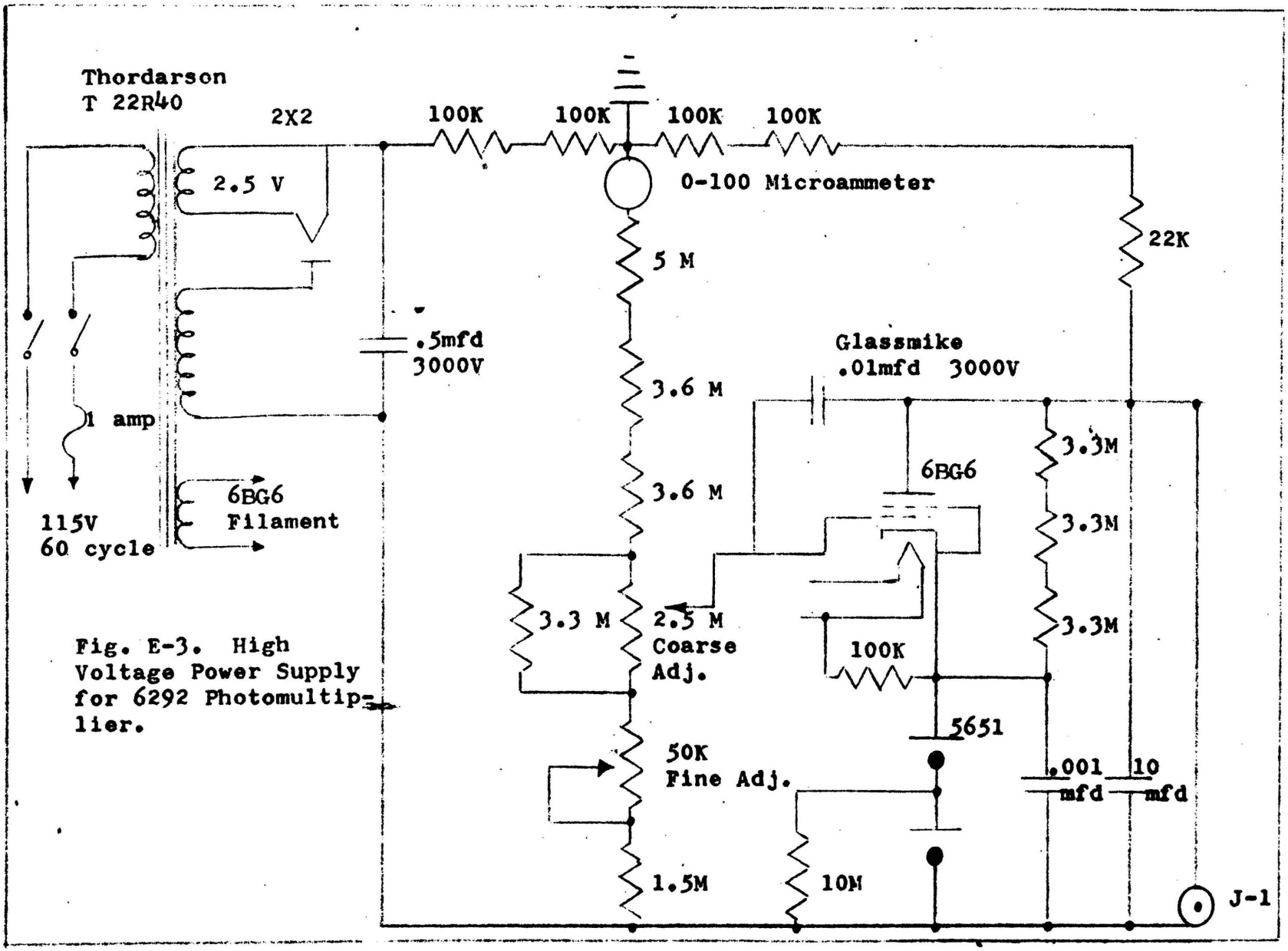


Fig. E-3. High Voltage Power Supply for 6292 Photomultiplier.

Appendix F

High Pass Filter

The filter network shown in Fig. F-1 was used to keep unwanted low frequency current from appearing on the oscilliscope. The direct current output of the photomultiplier tube was read on the microammeter shown.

If we let i be the current from the photomultiplier plate and i_2 be the current through the capacitor and resistor, the equation of the circuit may be written:

$$jwL(i - i_2) = (-j/wC + R)i_2$$

where w = 2 x 3.14 x frequency.

$$i_2R = i\ jwLR/(R + j(wL - 1/wC))$$

$$|i_2R| = i\ wLR/(R^2 + (wL - 1/wC)^2)^{\frac{1}{2}}$$

so we see that as w approaches infinity $|i_2R|$ approaches iR and as w approaches zero $|i_2R|$ approaches iw^2LCR which approaches zero. The values of L, C, and R shown in Fig. F-1 were chosen so as to be effective in discriminating against the sixty cycle component of the signal.

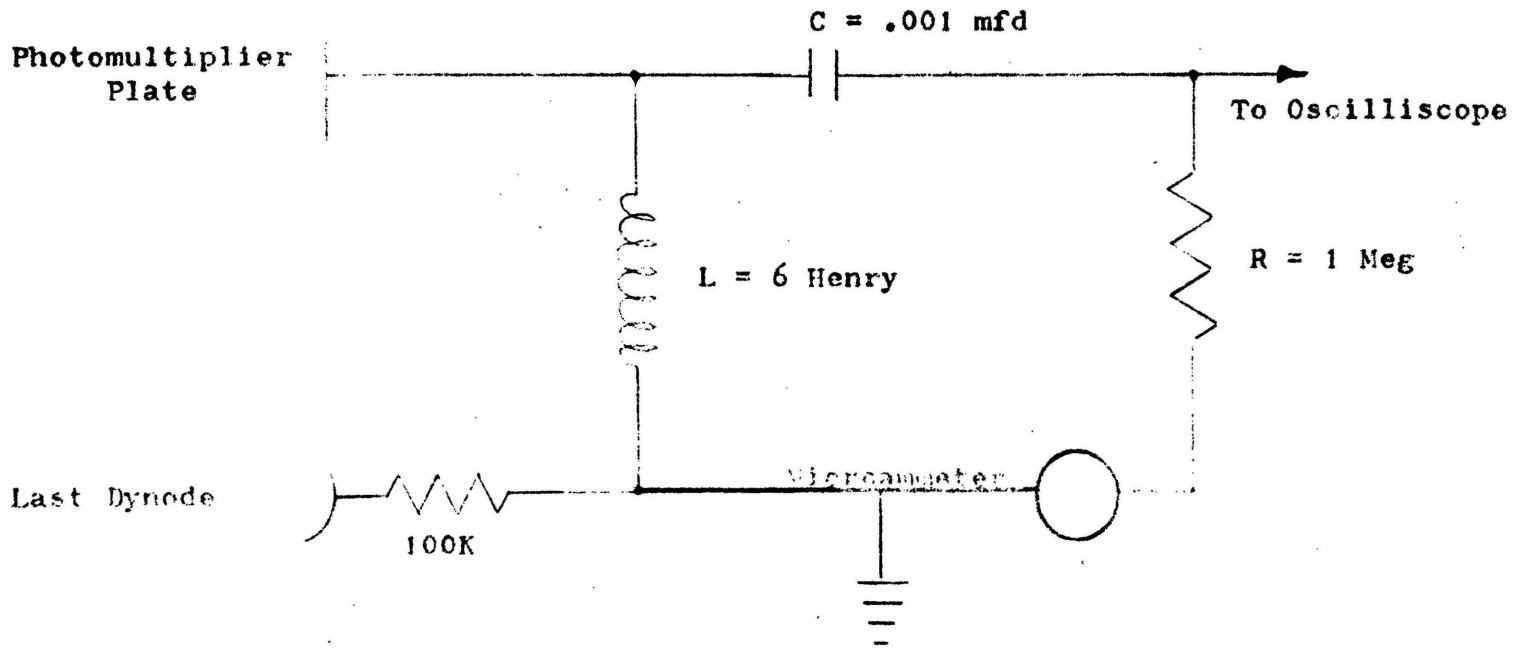


Fig. F-1. High Pass Filter

Appendix G

Additional Experimental Results

The oscilliscope traces shown in this appendix illustrate the variance of the signal with changes in grid bias and pulse length.

There is also shown in this appendix a sample data sheet showing the quantities measured for each run.

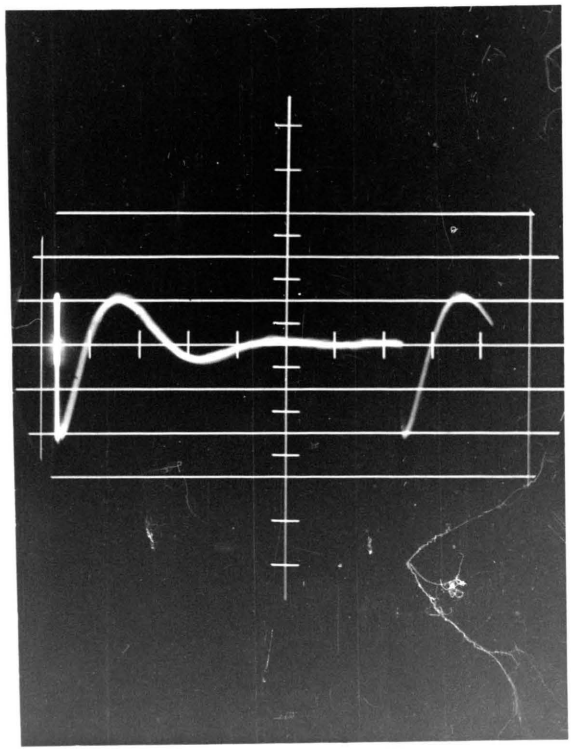


Fig. G-1. Signal Trace showing results at low grid bias, on the zinc oxide phosphor.

Pressure	5 micron
Grid Bias	-8.2 volts
Side Plate Voltage	-70 volts
Pulse Length	1 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Sensitivity	3 volts/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	78 microampere
Phosphor Plate Current	None

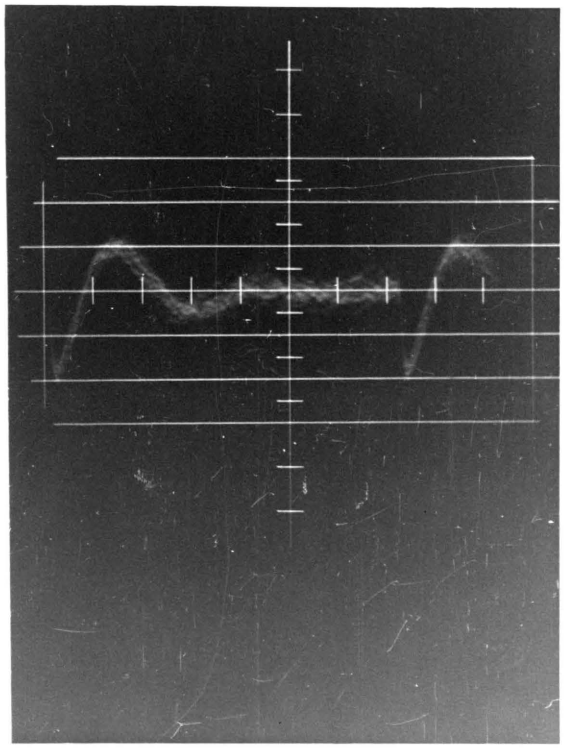


Fig. G-2. Signal trace showing results at high grid bias on the zinc oxide phosphor.

Pressure	47 micron
Grid Bias	-50 volts
Side Plate Voltage	+24 volts
Pulse Length	1 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Sensitivity	.3 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	15 microampere
Phosphor Plate Current	None

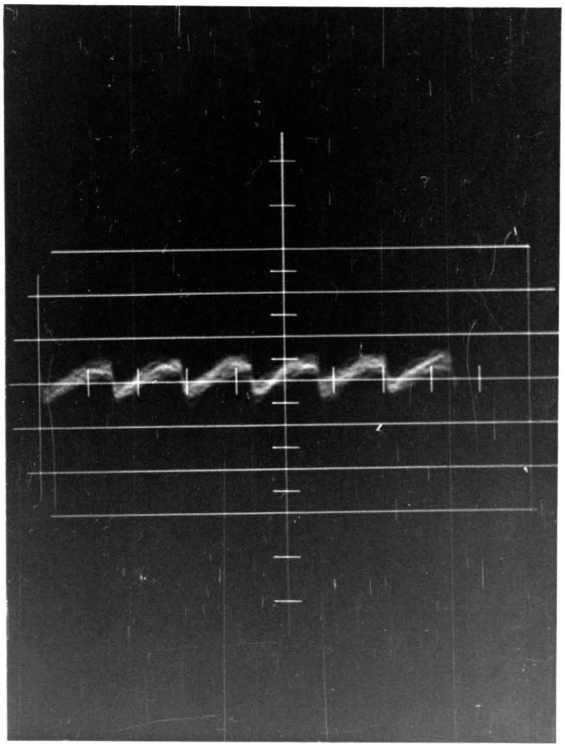


Fig. G-3. Signal trace showing effect of pulse length on the zinc oxide phosphor.

Pressure	42 micron
Grid Bias	-51 volts
Side Plate Voltage	0 volts
Pulse Length	1 microsecond
Pulse Frequency	5000/second
Oscilliscope Voltage Sensitivity	.3 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	9.6 microamperes
Phosphor Plate Current	None

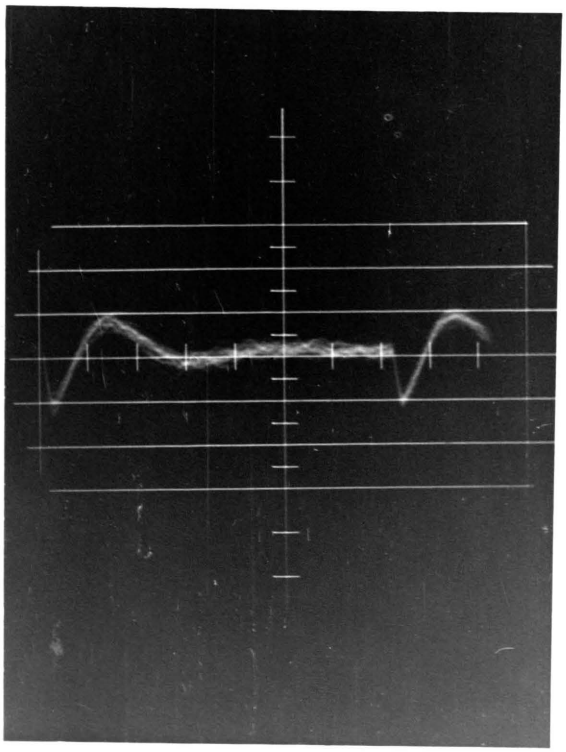


Fig. G-4. Signal trace showing effect of pulse length on zinc oxide phosphor.

Pressure	42 micron
Grid Bias	-51 volts
Side Plate Voltage	0 volts
Pulse Length	10 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Scale	.3 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	9.2 microampere
Phosphor Plate Current	None

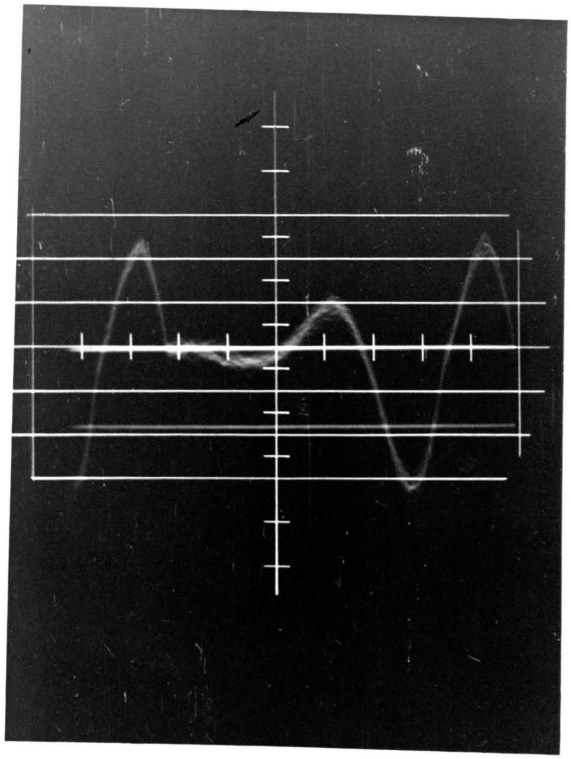


Fig. G-5. Signal trace showing effect of pulse length on zinc oxide phosphor.

Pressure	42 micron
Grid Bias	-51 volt
Side Plate Voltage	0 volts
Pulse Length	100 microsecond
Pulse Frequency	1000/second
Oscilliscope Voltage Sensitivity	.3 volt/cm
Oscilliscope Sweep Time	100 microsecond/cm
Photomultiplier Current	10.5 microampere
Phosphor Plate Current	None

Sample Data Sheet

	Run 1	Run 2	Run 3
Pressure			
Grid Bias			
Phosphor (Side) Plate Voltage			
Photomultiplier Voltage			
Pulse Length			
Pulse Frequency			
Oscilliscope Voltage Sensitivity			
Oscilliscope Sweep Time			
Photomultiplier Current			
Phosphor Plate Current			
Filament Current			
Annode Voltage			
Cathode Voltage			

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Vita

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In September, 1948, he enrolled in Buena Vista College and received a degree from that same institution in June, 1952. In September, 1952, he enrolled in the Graduate School of the Missouri School of Mines and Metallurgy as a candidate for the degree of Master of Science, Physics Major. Military service interrupted his formal education in June, 1953. In September, 1955, he returned to the Missouri School of Mines and Metallurgy to continue his education.